High-solid anaerobic digestion of sewage sludge: challenges and opportunities

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Abstract

High-solid anaerobic digestion (HSAD) of sewage sludge (SS) is a promising alternative to conventional anaerobic digestion (AD) as it reduces treatment volumes, transportation costs and energy consumption for heating and increases the fertilizing potential of SS. By centralizing sludge treatment, HSAD represents an opportunity to improve the energy balance of SS valorization compared to conventional AD. Nevertheless, HSAD of SS poses several challenges due to structural and rheological characteristics of dewatered SS and toxic compounds originating during (H_2S, NH_3, siloxanes, VOCs) and before (polyelectrolytes added prior to dewatering) the anaerobic treatment. This work critically reviews HSAD of SS with the objective to promote process enhancement and good practices for agricultural utilization of digestate. Advantages and drawbacks of HSAD are discussed in depth and practical solutions to solve critical issues at full scale are proposed.

Keywords

High-solid anaerobic digestion; sewage sludge; biogas; process inhibition; sludge rheology; agricultural recycling.

Abbreviations

AD	anaerobic digestion
ADS	anaerobically digested sludge
AOX	adsorbable organic halides
COD	chemical oxygen demand
cPAM	cationic polyacrylamide
CSTR	continuous stirred tank reactor
DEHP	di-2-etilesilftalato
DS	dry solids
DSS	dewatered sewage sludge
EPS	extracellular polymeric substances
FAN	free ammonia nitrogen
FBR	fluidized-bed reactor
FMS	fresh mixed sludge
GAC	granular activated carbon
HPUS	high-power ultrasound system
HSAD	high-solid anaerobic digestion
ISR	inoculum-to-substrate
LAS	linear alkylbenzene sulfonates

LCFA	long chained fatty acids
MAP	magnesium ammonium phosphate
NR	nutrient recovery
OFMSW	organic fraction of municipal solid waste
OLR	organic loading rate
OM	organic matter
OSC	organic silicon compounds
PAH	polycyclic aromatic hydrocarbons
PAM	polyacrylamide
PCB	polychlorinated biphenyl
PCB-DL	PCB-dioxin like
PCDD/F	polychlorinated dibenzodioxins and dibenzofurans
PCP	progressive cavity pump
PF	plug-flow
PSA	pressure swing adsorption
SAO	syntrophic acetate oxidation
SRB	sulfate reducing bacteria
SRT	solid retention time
SS	sewage sludge
TAN	total ammonium nitrogen
TFE	thin film evaporator
THP	thermal hydrolysis process

THS	thermal hydrolyzed sludge
TKN	total kjeldahl nitrogen
TOC	total organic carbon
TS	total solids
TSS	total suspended solids
VAH	volatile aromatic hydrocarbon
VFA	volatile fatty acids
VMS	volatile methyl siloxanes
VOC	volatile organic carbon
VRT	variable rate technology
VS	volatile solids
VSS	volatile suspended solids
WAS	waste activated sludge
WWTP	wastewater treatment plant

Nomenclature

γ	shear rate (s^{-1})
τ	shear stress (Pa s^{-1})
τ_0	yield stress (Pa s^{-1})
τ_c	critical shear stress (Pa s^{-1})
γ_c	critical shear rate (s^{-1})
η	apparent viscosity (Pa s)
η_∞	limit viscosity (Pa s)

K	fluid consistency coefficient (Pa s^n)
n	fluid behavior index
α_0	high shear viscosity rate (Bingham viscosity)

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1. Introduction

SS is largely produced in WWTPs during biological, chemical and physical treatments and is mainly composed of dewatered microbial biomass. Additionally, SS contains pathogens, heavy metals and other hazardous materials [1]. The amount of SS produced in European and developing countries is growing due to the increased demand for wastewater treatment [2]. The average SS production in Europe's top 13 producing countries between 2010 and 2017 ranged between 181 and 1850 Gg TS $year^{-1}$, resulting in an average specific production of 21 ± 4 kg TS $person^{-1}$ $year^{-1}$ (**Table 1**). Increase in SS production results in higher costs for pretreatment, transportation and disposal. As a result, the management of SS has become a major environmental and economic issue.

The applied sludge treatment and disposal methods include application to agricultural land (following AD, composting or chemical treatment), incineration, landfilling and recycling as building materials [3]. AD has the advantage of producing methane (CH_4) as the main

constituent of biogas (55-65 %), resulting in a source of renewable energy. The specific CH_4 production in conventional digesters treating sewage sludge typically ranges between 0.19 and 0.24 Nm^3 kg VS_{in}^{-1} [4] and depends on the SRT applied in the wastewater treatment line [5].

Table 1 - SS production and disposal methods in European countries between 2010 and 2017 [6].

	SS production (Gg DS year^{-1})	Specific SS production (kg DS person^{-1} year^{-1})	SS disposal method (%)			
			Agricultural use	*Composting*	*Incineration*	*Landfill + other*
Germany	1850	22.8	27.1	14.5	58.3	0.1
United Kingdom	1278	19.9	79.5	n.a.	19.8	0.7
Spain	1144	24.6	76.5	n.a.	4.4	19.1
Italy	1103	18.4	33.1	n.a.	3.8	63.1
France	1060	16.1	44.3	30.4	19.2	6.1
Poland	550	14.5	20.1	6.3	12.8	60.8
The Netherlands	348	20.7	0	0.2	99.4	0.4
Austria	252	29.5	16.8	18.7	48.7	15.8
Czech Republic	227	21.6	41.0	42.6	5.0	11.4
Sweden	203	21.1	28.5	32.7	1.4	37.4
Switzerland	195	24	0	0	96.8	3.2
Hungary	186	18.7	9.8	46.1	22.0	22.1
Portugal	181	17.3	43.6	n.a.	0	56.4

n.a. = not available.

Additionally, AD reduces the amount of sludge solids for final disposal, stabilizes the sludge, destroys the pathogens and limits odor emissions [7,8]. The main drawbacks of AD of SS are: 1) the low reaction rates (due to the slow hydrolysis of bacterial aggregates), resulting in large reaction volumes and high investment costs for the digesters; 2) process vulnerability and low resilience to inhibitor (e.g. ammonia) accumulation; 3) production of hydrogen sulfide (H_2S) and volatile silicon compounds, which hamper biogas production and utilization. Other disadvantages are the high buffer requirement for pH control, poor efficiency for the treatment of diluted waste and increased concentration of heavy metals in the ADS [8,9].

TS content is a key parameter impacting the overall digestion performance. Conventional AD of SS is carried out with a TS content between 2 % and 6 %, as pumping and mixing of the sludge becomes challenging at higher TS content [8]. However, TS content can be increased up to 25 % for 1) reducing the storage area within the WWTP and 2) reducing the cost of transportation. Increase in TS levels results in higher digester capacity and reduces water addition to the feed

substrate [10]. On the other hand, HSAD results in the accumulation of inhibitory metabolites such as H_2S, FAN and LCFA, which can disrupt or slow down methanogenic activity [11–13]. Additionally, DSS possesses high viscosity, which can severely limit mixing and pumping operations in the digester.

Previous reviews on HSAD focused on organic substrates other than SS, such as OFMSW, animal manure and hydrophobic matrixes such as lignocellulosic materials, for which operational TS content can be as high as 40 % [14]. Such high TS levels are inapplicable when DSS is used as a feed substrate due to its peculiar rheological properties [15]. The anaerobic treatment of DSS with TS > 6 % requires specific technologies and techniques for sludge pumping and transportation and for ADS distribution on the agricultural fields as well as the optimization of anaerobic digester design, process control and specific pretreatments to improve sludge biodegradability.

The aim of this review is to provide a comprehensive tool for understanding the complexity and technological challenges of HSAD of SS and provide hints for a viable and economically advantageous full-scale application of the process. The environmental factors and process parameters influencing the HSAD of SS are critically analyzed. Additionally, the effects and technological advances regarding the application of high-solid ADS to the agricultural lands are discussed. Research needs are highlighted to promote progress and fill the literature gaps in this field.

2. Overview of HSAD of SS

Sludge treatment in WWTPs usually includes thickening and dewatering, which separate the solid and liquid components of the sludge to be easily handled for final disposal. Polyelectrolytes are often added to enhance floc adhesion and improve dewatering efficiency [16]. Anaerobic digesters generally require huge operating volumes due to the high water-content of the treated sludge, resulting in insufficient biogas production to sustain the energy demand of the WWTP [17]. Consequently, conventional AD is not always foreseen in small WWTPs and highly urbanized areas with limited space [18]. In the recent decades, small towns and cities have shown an increasing trend to merge together for a more effective and convenient utilization of the infrastructures, including sewage and waste treatment facilities [19]. Centralized AD plants collecting DSS from different WWTPs can serve as hubs for AD and strongly reduce the operational and investment costs for sludge treatment. Digesting sludge with high TS concentration allows to reduce the treatment volumes while maintaining the same VS loading rate, which significantly reduces power consumption for heating as well as transportation costs. As a result, centralizing SS valorization results in energy-neutral or even net-energy-positive AD plants.

However, the high TS content of DSS raises several challenges for the digestion process:

1) The hydrolytic stage is typically the rate-limiting step of AD, as secondary sludge is rich in bacterial cells difficult to digest [20]. The polyelectrolytes added as flocculants further limit the hydrolysis rate and have been recently shown to deteriorate methane production [21]. Thermal and ultrasonic pretreatments, chemical conditioning and cavitation are usually applied to improve sewage sludge biodegradability and enhance hydrolysis [7].

However, the application of these treatments to DSS is not always possible due to rheological issues.

2) The HSAD of SS leads to high ammonia accumulation due to the anaerobic degradation of proteinaceous material. SS is rich in proteins, being the protein content between 12 % and 29 % of TS for primary sludge and between 25 % and 50 % of TS for secondary sludge [22]. The risk of ammonia inhibition increases considerably under thermophilic conditions, as the fraction of FAN (the most toxic form of ammonia) increases with temperature [23]. As a result, ammonia removal could be necessary to avoid the disruption of the anaerobic process.

3) SS is a pseudoplastic fluid and shows yield stress and viscoelastic behavior when concentrated or dewatered [24]. Rheological and CFD studies and installation of specific devices might be required for adequate pumping and mixing at high TS concentrations, which might lead to considerably higher investment and operational costs compared to conventional AD systems.

4) Biogas from AD of SS is commonly used in 1) boilers for heat generation, 2) combustion engines for the combined production of heat and electricity and 3) upgrading plants for the conversion to biomethane. These technologies usually require purification pretreatments prior to biogas utilization [25]. HSAD of SS produces a biogas rich in H_2S, VOC and siloxanes and requires a rigorous biogas purification.

Besides these difficulties, ADS has a great potential as soil amendment and/or fertilizer [26]. Generally, the composition of ADS includes 30-55 % of stabilized OM, up to 3 % of total

nitrogen, 0.7-1.5 % of total phosphorus, 0.7 % of total potassium and various levels of magnesium, sulfur and heavy metal ions. The heat value of dry sludge ranges between 12000 and 15000 kJ kg^{-1} [27].

The recycling of ADS for agricultural purposes is one of the most sustainable option for its disposal as it embraces the principles of circular economy [28]. In Europe, each member state has issued a national legislation based on the European directive 86/278/EEC which disciplines the use of ADS in agriculture and, in some cases, has set more stringent limits for pathogen, metals and organic micropollutants in biosolids and soils [3]. In the last 10 years, HSAD of SS has been increasingly studied at different scales with the main objectives to enhance methane production and investigate process inhibition. A comprehensive list of applications on HSAD of SS in continuous and batch bioreactors is provided in **Table 2** and **Table 3**, respectively.

Substrate	Volume (L)	TS (%)	T (°C)	pH	Pretreatment/ enhancement strategy	Mixing	SRT (d)	OLR (kg VS m^{-3} d^{-1})	CH_4 yield (L kg VS_{added}^{-1})	TAN, FAN (g L^{-1})	VFA (g L^{-1})	Reference
DSS	20 [a]	17.5	35	7.5-7.8	None	Yes	~ 30	2.7-4.5	115-163	< 0.2 (FAN)	≤ 0.6	[1]
DSS	6 [a]	10-20	35 ± 1	7.3-8.3	None	Yes	4-59	2.0-12.8	120-270	≤ 4 (TAN) ≤ 0.8 (FAN)	< 4	[18]
DSS	3-10 [a]	5-10	35°C (mesophilic) 55°C (thermophilic)	6-8	None	Yes	>28-84	1.25-3.3 [c]	-	< 4 (TAN) < 0.6 (FAN)	< 0.2 (mesophilic) ≤ 10 (thermophilic)	[19]
DSS	3 [a]	10	35	6.5-8	None	Yes	n.a.	1.0-4.2	40-620 (SBP [d])	1-5 (TAN)	≤ 10	[29]
DSS	9	15	35 ± 1	7.9-8	None	Yes	20	n.a.	9.8-11.9 [g]	≤ 6 (TAN) < 0.6 (FAN)	≤ 18	[30]
DSS (90%) + OFMSW co-products	13.5*10^6 [b]	10.7	55	7.86	None	Yes	44-71	1.5-2.3	186-218	3.5-3.8 (TAN)	n.a.	[31]
DSS	1.5*10^6	10-12	mesophilic	n.a.	Cambi's THP	Yes	17	3.7	~270	n.a.	n.a.	[32]
DSS	2.45*10^6	24	mesophilic	n.a.	Exelys THP	Yes	15	3.1-4.2	~300	n.a.	n.a.	[33]
DSS	450 [a] 500 [b]	15	35 ± 2	7.8-7.9	THP (70°C) for 30 min	Yes	15-22	3.4-5.6	333–408 (SBP [d])	< 2.5 (TAN) ≤ 0.2 (FAN)	< 0.45	[17]
DSS	0.5 (5-10% TS) 2 (16%TS)	5-16	35 ± 1	n.a.	THP (80°C, 24 h) for 5%TS only	No	n.a.	0.5-2	200-300	< 60 g kg TS^{-1} (TAN)	n.a.	[34]
DSS + cattle manure (VS ratio: 3/7)	7-9 [a]	16.0 (DSS), 17.5 (cattle manure)	35 ± 1	9	Co-digestion	Yes	12-20	n.a.	~ 190 [e]	n.a.	53-68 [f]	[35]

Table 2 – Operational conditions and performances of HSAD of SS in continuous bioreactors.

n.a. = not available; [a] working volume; [b] total volume; [c] calculated based on volumetric loading rate; [d] SBP = specific biogas production (L_{biogas} kg VS_{added}^{-1} d^{-1}); [e] calculated from L_{CH_4} kg TS_{added}^{-1} d^{-1}; [f] reported as g VFA kg TS^{-1}; [g] reported as L_{biogas} d^{-1};

Volume (L)	TS (%)	T (°C)	pH	Pretreatment/ enhancement strategy	Mixing	Digestion time (d)	Cumulative CH_4 yield (L kg VS_{added}^{-1})	TAN, FAN production (g L^{-1})	VFA yield (g L^{-1})	Reference
0.25 [b]	10-16	35 ± 2	7-8.5	None	Yes	38-46	115-121 [d]	≤ 4 (TAN) ≤ 1 (FAN)	≤ 8 (as HAc)	[36]
0.2 [a] 0.25 [b]	~ 10 [c]	35 ± 1	7.6-7.8	Fe^0 addition (0-20 g L^{-1}) during digestion	Yes	22	146-215 [e]	3.64-4.07 (TAN)	≤ 3	[37]
2.5 [b]	6-15	35 ± 2	7-8	None	No	60	20-190	< 2.5 (TAN) < 0.25 (FAN)	< 0.4	[38]
0.5 [b]	16.7	37	n.a.	THP (60-90°C) for 1-72 h THP (120-180°C) for 15-180 min	Yes	28	940-1070 [f]	n.a.	~ 0.03 [e]	[39]
0.5 [b]	15	35 ± 2	7.6-8.1	THP (60-80°C) for 30 min	Yes	20-24	93-108 [d]	215-272	< 750	[17]

Table 3 – Applications of HSAD of SS in batch bioreactors.

n.a. = not available; [a] working volume; [b] total volume; [c] reported as TSS; [d] reported as L_{biogas} kg VS_{added}^{-1}; [e] reported as L kg VSS^{-1}; [f] reported as L_{biogas} g $VS_{removed}^{-1}$; [g] reported as g VFA kg TS^{-1}.

3. Process challenges and solutions

3.1. Pretreatments to enhance the anaerobic biodegradability of DSS

Despite the previously mentioned benefits deriving from the anaerobic treatment of SS, AD is generally characterized by long retention times (≥ 20 d) and low VS degradation (30-50 %) [9]. The scientific community has identified the cause of these limits in the slow hydrolysis of the cell aggregates composing secondary sludge [40,41].

Indeed, besides the large amount of water (> 75 %), SS mainly contains microbial aggregates (flocs), constituted by microorganisms held together by EPS, which are generally composed of proteins, polysaccharides and humic-like substances. EPS, deriving from the microbial metabolism and lysis or adsorbed from the bulk solution, create a three-dimensional matrix bound to the surface of the cells, thus generating a shield that protects the microorganisms contained in the aggregate. Specifically, EPS avoid the rupture and the lysis of the cells, increase

the strength, and decrease the dewaterability and biodegradability of the flocs [42,43]. In addition, each microorganism is protected by a cell membrane composed of a phospholipid bilayer with embedded proteins that acts as a physio-chemical barrier to direct AD [8].

Moreover, cPAM, which may limit the hydrolysis rate and decrease methane production, is generally added within the range of 2.5-10 g kg TS^{-1} during the thickening and dehydration treatments [44,45]. cPAM are used with the aim of aggregating the flocs and the other particles present in the sludge, through charge neutralization and interparticle bridging, increasing the dewaterability and decreasing the transport costs of DSS.

With the aim of destroying microbial aggregates and cells before AD of the SS, in the last decade, many studies were focused on mechanical, thermal and chemical pretreatments. As a consequence of these pretreatments, the above described slowly biodegradable biomasses are converted to lower molecular weight and faster biodegradable compounds, thus increasing the hydrolysis rate, the VS conversion efficiency and the consequential bio-methane production of HSAD.

3.1.1. Thermal pretreatments

High temperature pretreatment sterilizes the sludge and dissolves the EPS both inside and on the surface of the flocs, thus disintegrating the flocs structure and increasing both the bioavailability of the materials that compose the cells and the dewaterability of SS [39,46]. In addition, the cell destruction due to pressure differences under the thermal process may contribute to a further increase of hydrolysis rate [32]. The first full scale plant aimed at thermally hydrolyzing DSS before HSAD was the Cambi process, which was designed as an alternative to the Porteous and

Zimpro processes [47]. The main goal of these two processes was the increase in dewaterability of the sludges, but they were characterized by high costs, odor release and high strength COD liquor production [48]. Nonetheless, at the high operating temperatures (at least 30 min at 200 °C) characterizing the Porteous and Zimpro processes, the Maillard reactions may quickly take place [49]. As a result, melanoidins that are difficult to degrade or even inhibit the degradation of other organic products are produced, thus hindering any biological sludge treatment [50].

In particular, the first Cambi's THP was installed in 1995 at the WWTP of Hamar, Norway [32]. The Cambi's THP is designed to operate with DSS having a TS content > 16 %. It mainly consists of three units (**Figure 1A**): the pulper, hydrolysis batchwise reactors and the flash tank. The TS concentration of the sludge in the pulper is decreased to 14.5-16.5 % using the steam recirculated from the hydrolysis reactors and the flash tank, thus allowing a preheating of the sludge to 70 °C. Later, the sludge is pumped to the reactor tank where it is heated to 160-180 °C under a pressure of approximately 5-6 bar.

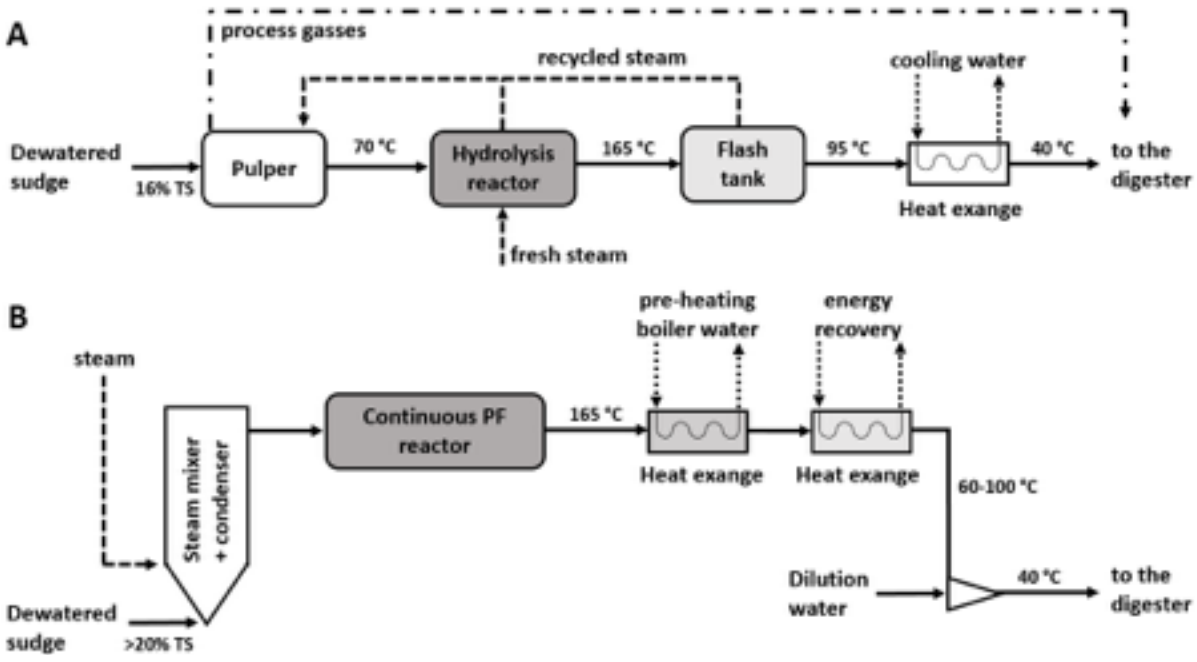


Figure 1 - Cambi™ (a) and Exelys™ (b) systems for the thermal hydrolysis pretreatment.

After 20-30 min hydrolyzing treatment, the sterilized sludge is pumped to the flash tank, where the rupture of the cells occurs due to the fast pressure drop [51]. Generally, the sludge from the flash tank is too hot to feed a mesophilic digester and it needs to be cooled to 42-44 °C by heat transfer. In this way, 60 % of the VS can be converted to biogas during HSAD [52]. Moreover, unlike Porteous and Zimpro processes, the Cambi's process does not release odors because the steam released both from the hydrolysis reactor and the flash tank is recycled to the pulper and the process gases resulting from the pulper are sent to the digester.

Another commercial alternative to the Cambi's process is represented by the Exelys system, developed by Kruger Inc., a subsidiary of Veolia Water. Exelys is a continuous PF system which can operate with DSS containing > 20 %TS. In particular, the raw DSS and the steam are continuously fed to a mixing and condensing tank and to the PF hydrolysis reactor maintained at 165 °C and 9 bar (**Figure 1B**). Afterwards, the hydrolyzed sludge is cooled, diluted and treated by HSAD. This process seems to require more steam per ton of dry solids than the Cambi's system because there is no recycling of the steam. However, since Exelys system can operate with higher TS concentrations, the steam requirements, equal to 1.31 kg steam per kg TS, is reduced by 30 % with respect to the Cambi's process [33]. Specifically, when the sludge enters the flash tank, the water contained in the flocs and inside the cells partially vaporizes, resulting in their structure damaging. Thus, the presence of the flash tank seems to be important for a sufficient hydrolysis of the sludge [51,53–55]. On the other hand, some studies highlighted that flash tank may not result to valuable differences in terms of COD and VSS solubilization

[33,56]. In addition, both Exelys and Cambi's treatments lead to sludge sanitation, reduction of the viscosity, enhancement of sludge handling and positive energy balance compared to conventional sludge treatment, since the energy consumed with these processes may be covered by excess methane production [32].

Besides the two processed previously described, there are other commercial thermal hydrolysis pretreatments. Even if these processes are characterized by different operating schemes, the operation parameters and the performances are similar (**Table 4**).

The above-mentioned processes, generally carried out at high temperatures (> 140 °C), are characterized by high energy consumption and place strict demands regarding devices and high costs, which may offset the benefits of these pretreatments [57]. With the aim of overcoming these drawbacks, thermal pretreatments characterized by a lower operational temperature (50–90 °C) have been tested at laboratory scale. The results show that also low-temperature thermal treatment can hydrolyze the sludge, thus increasing the methane production and decreasing the viscosity [17,34,58,59]. However, when low-temperature thermal treatments are compared with high-temperature thermal treatments by using the same sludge and the same reactors, laboratory study highlighted that the best results are obtained with operational temperatures in the ranges of 140-160 °C [39]. Specifically, Xue et al. [39] reported that, with respect to raw or pretreated DSS (16.7 %TS) at temperatures < 120 °C, the biogas production increased by 6–16 % and the SRT could be reduced from 18–20 d to 12–14 d after a sludge pretreatment carried out at 140–160 °C. However, another study demonstrated that even if the organic content became more solubilized when the pretreatment temperature is increased within the range of 140–165 °C, with soluble COD increasing from 32 to 45 mg L^{-1}, this additional solubilized material was not degradable

due to the formation of melanoidins, which are not biodegradable [60]. In addition, this temperature would also reduce the ammonification of proteins contained in the sludge, thus decreasing the amount of ammonia, a methanogenesis inhibitor, in the pretreated sludge fed into the HSAD [61].

Table 4 - Comparison between the available commercial thermal hydrolysis systems.

Process	**Cambi**	**Exelys**	**Lysotherm**	**HCHS**	**Turbotec**	**Aqualysis**	
Company	Cambi	Veolia Water	Eliquo STULZ	Haarslev Industries	Sustec	Aqualogy	te
Batch	Yes	No	No	No	No	No	No
TS in feed	16-18 %	> 22 %	2-12 %	17-22 %	10-12 %	16 %	5-15 %
			Operating conditions of the hydrolysis reactor				
T (°C)	150-180	165	140-175	150-170	140-175	165-180	220
P (bar)	5-6	9	5-10	7-8	4-6	7-10	12-14
HRT (min)	20-30	30	30-60	20	30-70	15-30	< 5
Flash tanks	Yes	No	Yes	Yes	No	Yes	Yes[a]
Heat exchangers	No	Yes	Yes	No	Yes	No	No
Pumps	Yes	Yes	Yes	No	Yes	Yes	No
				Effects on subsequent steps			
Biogas production increase[b]	100-150 %	30-50 %	20-50 %	-	35 %	30 %	30 %
VS abatement	60-70 %	25-35 %	-	-	30 %	30 %	-
TS after HSAD sludge dewatering	30-40 %	22-30 %	25-35 %	-	> 30 %	-	-

[a] There are two flash tanks. [b] It has been considered with respect to the HSAD without pretreatment.

3.1.2. Physical pretreatments

Physical pretreatments, such as sonication, lysis-centrifuge, collision and high-pressure homogenizer, were extensively studied to increase the efficiency of AD since they disintegrate flocs, cells and other organic particles. Thus, these pretreatments lead to a release of the compounds contained in the cells and the solid fragments in the effluents are characterized by higher specific surface area. The latter increases the interactions occurring between solids and bacteria enzymes, thus enhancing the AD process [62–64]. The mechanism of particle destruction during a sonication pretreatment is mainly attributed to the cavitation phenomena and the efficiency depends on the ultrasound source and sludge properties. In particular, ultrasound wave propagation in a medium is characterized by energy losses due to attenuation, adsorption and dissipation. Specifically, Abramov [65] evaluated the sound intensity at the distance z from the source (I_z) as follows:

(1)

where I_0 is the sound intensity at the source and ϕ is the sound absorption coefficient, which depends on the medium viscosity. Consequently, an increase of solid concentration leads to greater energy losses and reduces ultrasound intensity in areas far from the source, thus hindering the development of cavitation bubbles in remote regions. Indeed, it has been demonstrated that the optimal TS concentration is the range of 2.3-3.2 % [66,67]. Similarly, the HPUS developed by ULTRAWAVES GmbH, is designed to work with TS contents lower than 6 % (https://ultrawaves.de/technology/ultrawaves-high-power-ultrasound-systems). As a consequence, DSS cannot undergo to sonication pretreatments as it is generally characterized by a higher TS

content. For the same reasons, lysis-centrifuge, collision and high-pressure homogenizer cannot be applied to this kind of waste.

3.1.3. Chemical pretreatments

Ozonation of excess sludge is an effective and well documented AD pretreatment, due to both radical and ionic attack of dissolved ozone molecules against the OM present in the solution [68,69]. In particular, the reactions between ozone and polysaccharides, proteins and lipids lead to the production of smaller molecular-weight compounds, so that the membrane of the cells is destroyed, spilling the cell cytoplasm [70]. Indeed, an ozone dose in the range of 0.05-0.5 g O_3 g TS^{-1} results in a higher VSS removal efficiency of 81 % together with a higher methane production [71,72]. As an example, a full-scale test lasting two years demonstrated that the ozonation pretreatment can lead to a 30 % increase of methane production during the AD of SS [73]. Additionally, the occurrence of radical reaction leads to the oxidation of recalcitrant compounds, thus decreasing the risk associated with a sludge reuse in agriculture [74–77]. Up to date, there is no information in scientific literature regarding the adoption of ozonation as a pretreatment for HSAD of SS. However, this technology has been successfully adopted for the HSAD pretreatment of OFMSW [78].

Alkaline conditioning has been largely studied as pretreatment before AD of municipal excess sludge [79–82]. Indeed, the addition of bases, such as Na(OH) or $Ca(OH)_2$, leads to a fast pH variation, which changes the osmotic pressure of the cells contained in the sludge resulting in EPS solubilization and cell lysis. In particular, the addition of different NaOH doses (0-80 mg NaOH g TS^{-1}) to DSS containing 22.9 %TS was investigated as a pretreatment before HSAD

[83]. The results show that the higher is the NaOH dose, the higher is the amount of dissolved carbohydrates, proteins, COD and ammonia. These results are in agreement with a previous study [84] focused on the VS solubilization and COD increase during a thermal-alkaline pretreatment of DSS (TS = 10 %). Indeed, the COD solubilization and VS hydrolysis rates were expressed as pseudo first-order rate expressions:

(2)

(3)

where k is the first order hydrolysis rate constant (h^{-1}), COD_s represents the soluble COD (mg L^{-1}), COD_∞ is maximum theoretical soluble COD and represents the d(COD)/d(VS) correlation parameter. In particular, Vlyssides and Karlis [84], on the basis of the results deriving from an experimental campaign consisting of 20 hydrolysis experiment carried at different temperatures (50-90 °C) and pH (8-11), obtained the expression of k and COD_∞ as a function of both temperature and pH:

(4)

(5)

However, the best results in term of cumulative methane production were observed when only 20 g NaOH kg TS^{-1} were added during the pretreatment [83]. Indeed, the higher NaOH doses during the pretreatment lead to an increase of Na^+ concentrations in the sludge, which may inhibit the activity of acetoclastic methanogens and limit the reuse of the digested sludge in agriculture [85].

Recently, it has been reported that the addition of 10 g L^{-1} iron scrap to a DSS containing almost 10 %TS may lead to an increase of both methane yield and VS removal during HSAD [37]. In

particular, when rusty iron scraps (8 mm x 4 mm x 0.5 mm) were used, methane yield and VS removal increased by 29.6 % and 27.3 %, respectively. These results are ascribed to the combined action of Fe^0 and Fe(III) oxides constituting the rusty iron scraps. Indeed, in addition to decreasing the oxidative-reductive potential [86], Fe^0 powder may increase the activity of the enzymes associated with hydrolysis–acidification by 2–34 times [87]. On the other hand, Fe(III) oxides prompted a microbial iron reduction to decompose complex matters of the sludge, which behave like an electron donor, thus accelerating the hydrolysis rate [37].

3.2.Ammonia toxicity on HSAD of SS and remedial actions

3.2.1. Overview of ammonia toxicity on AD of SS

Ammonia exists in water in ionized (NH_4^+) and unionized gaseous form (NH_3). TAN is defined as the sum of ammonium nitrogen ($N\text{-}NH_4^+$) and FAN ($N\text{-}NH_3$). The relative concentration of each component depends on pH and temperature, as the increase of these two parameters leads to a higher FAN fraction according to the following relationship [88]:

(6)

where FAN and TAN are expressed as mg L^{-1} and temperature (T) as K. Fernandes et al. [89] showed that FAN accounted for less than 1 % of TAN in anaerobic digesters operated at pH 7 and 35 °C, while 10 % FAN was measured at pH 8. In another study, Kayhanian [90] showed that FAN concentrations at thermophilic temperatures (55 °C) was six times higher than that observed under mesophilic conditions at the same pH. **Eq. 6** should be applied only in the case of diluted systems, which show a chemical behavior similar to that of an ideal pure water-ammonia solution. In the case of high TS solutions such as DSS, the effect of ionic strength on

the chemical activity of the ions should be accounted and an activity coefficient can be applied as a correction factor into the ideal equilibrium equation [91].

During AD, TAN originates from the hydrolysis of organic nitrogen in the substrate, i.e. proteins, amino acids and urea. Theoretically, the quantity of ammonia generated by AD of biodegradable organic substrate can be estimated using the modified Buswell's equation [92] simplified by disregarding sulfur:

(7)

Assuming $C_5H_7O_2N$ as a representative formula for secondary sludge, about 0.15 g of NH_3 are generated from the anaerobic degradation of 1 g VS of sludge. However, the fraction of total nitrogen in the substrate converted to ammonia is usually in the range of 30-50 %, depending on the composition and degradability the organic substrate and on the process temperature [93,94].

TAN is a micronutrient, being required for microbial growth, and acts as a buffer in the system by counteracting acidification due to VFA production. TAN concentration as low as 500 mg L^{-1} was shown to have detrimental effects on AD, i.e. low methane yield, loss of biomass and reduced methanogenic activity, due to insufficiency of nitrogen as a nutrient [95]. Conversely, TAN concentrations above 1500 mg L^{-1} are commonly reported to inhibit AD [91]. Ammonia inhibition proceeds via several mechanisms, i.e. proton imbalance, potassium deficiency, change of intracellular pH, increase of energy requirement and inhibition of enzymatic reactions [11,96]. FAN is considered the most toxic ammonia fraction as it easily diffuses into the cell membrane. Compared to hydrolyzers and acetogens, methanogens are more sensitive to several types of environmental stress, such as VFA or ammonia overload [90]. In particular, acetoclastic

methanogens (which produce methane by consuming acetate or acetic acid) possess a lower tolerance to ammonia compared to hydrogenotrophic methanogens and SAO bacteria [23]. Ammonia inhibition of methanogenesis also results in a significative pH drop in the system as methanogens would not be able to consume the VFA produced by the acetogens. The deterioration of the digestion performance at high TAN concentrations can be also attributed to a decrease in bacterial diversity, which may detrimentally impact the hydrolysis of carbohydrates and proteins and limit the supply of acetate to methanogens [30]. Increase in TAN concentration and OLR has been shown to promote SAO coupled to hydrogenotrophic methanogenesis [97], although stable acetoclastic methanogenic communities have also been observed under ammonium stress (5-6 g TAN L^{-1}) [30].

TAN concentration in the digester liquor of a municipal WWTP sludge line commonly ranges between 1000 and 2000 mg L^{-1}, while pH is about 7.5-8.6. This results in FAN levels between 30 and 560 mg L^{-1} at 33 °C [98]. Literature studies report TAN and FAN inhibitory limits (50% inhibition) for methanogens ranging from 1100 to 11800 mg L^{-1} and from 27 to 1450 mg L^{-1}, respectively [91]. The causes of this large variability have been attributed to the use of different microbial communities, temperatures, pH and ionic strength values, acclimation strategies and FAN calculation models.

3.2.2. Influence of ammonia on HSAD of SS

Digesting sludge with TS concentration between 10 % and 20 % under mesophilic conditions generates TAN and FAN levels from 2000 to 4000 mg L^{-1} and from 200 to 800 mg L^{-1}, respectively [18]. Dewatering leads to an exponential increase in sludge viscosity and decrease in

the diffusive coefficient, which results in a blocked mass transfer and leads to VFA and FAN accumulation. This can further reduce methanogenic activity and rapidly lead to AD failure [38].

In order to limit ammonia inhibition, the OLR applied for HSAD of SS cannot be too high. As shown in **Table 2**, OLRs applied for HSAD of SS mostly falls within the recommended range for conventional AD, i.e. 0.5-7 kg VS m^{-3} d^{-1} [99].

Hidaka et al. [19] showed that DSS with 7.5 %TS could not be successfully digested when TAN concentration exceeded 2000 mg L^{-1} under thermophilic conditions, whereas stable HSAD could be maintained at 10 %TS and TAN concentrations between 3000 and 4000 mg L^{-1} under mesophilic conditions. In contrast, Giordano et al. [31] observed no inhibitory effects on methanogenic activity in full-scale thermophilic digesters co-digesting DSS with 11 %TS at 3.5–3.8 g TAN per kg of ADS. Additionally, the authors reported that the high TAN levels occurring in the digesters likely inhibited SRB activity, reducing H_2S production and, thus, the oxygen flow rate required for H_2S oxidation in the headspace.

Capson-Tojo et al. [91] explained that the lower TAN inhibition thresholds often reported for thermophilic AD compared to mesophilic AD are a consequence of the much higher FAN/TAN ratios occurring at thermophilic conditions. Using a clustering approach, the authors revealed that, in fact, thermophilic methanogens possess higher resilience and inhibitory thresholds than mesophilic ones, which agrees with thermophilic archaeal communities being more exposed and adapted to elevated FAN concentrations.

Hydrolysis can also be severely inhibited due to high TAN concentrations during HSAD. Dai et al. [30] observed that TAN levels as high as 6000 mg L^{-1} dramatically reduced the population of

protein-degrading bacteria during mesophilic digestion of DSS (20 %TS), while carbohydrates degradation was less affected. Based on these results, protein degradation seems more vulnerable to ammonium stress than carbohydrate degradation. The inhibition of the hydrolytic step can substantially reduce methanogenic activity by limiting acetate availability.

3.2.3. Remedial actions for ammonia disinhibition during HSAD

Physical-chemical methods, including gas stripping, dilution, chemical precipitation, adsorption and mixing, and biological methods such as biomass acclimation, bioaugmentation and immobilization have been tested to reduce ammonia toxicity during AD of different organic substrates [96,100]. As described in Section 3.2.2, HSAD of SS can generate ammonia concentrations above the inhibitory levels due to the anaerobic degradation of concentrated proteinaceous material. Nevertheless, a limited number of applications on ammonia removal during HSAD exists in the literature.

3.2.3.1.Ammonia stripping

Gas stripping has been successfully applied for ammonia disinhibition during HSAD of SS. Stripping of ammonia is conventionally performed in heated packed column reactors on the liquid fraction obtained from sludge dewatering [101,102], as direct DSS feeding would result in rapid clogging of the column. However, sludge dewatering is energy-intensive and requires the addition of polyelectrolytes for effective solid-liquid separation, which significantly increases the costs for ammonia removal and might affect methane production (see Section 3.3).

TFE has been recently proposed as an alternative to conventional packed columns when treating DSS, as this technology does not require solid/liquid separation and avoids clogging issues.

Costamagna et al. [103] applied a TFE system for the simultaneous stripping of NH_3 and CO_2 during HSAD of SS (12.6 %TS), which was recirculated directly from the digester to the TFE unit. TAN removal was performed by exposing a thin layer of ADS to a high-rate (5 $Nm^3\ h^{-1}$) uprising biogas flow continuously circulated from the bottom of an adsorption column, while diluted sulfuric acid was fed from the top. Results showed that 19.3 g $N\text{-}NH_4^+$ per kg TS could be stripped from the ADS of a thermophilic digester performing HSAD during 180 days of operation.

3.2.3.2.Struvite precipitation

Crystallization and separation of MAP, also known as struvite, is an efficient method to efficiently recover nitrogen and phosphorus from the supernatant of anaerobic digesters [104]. MAP precipitation from AD supernatants is commonly performed in FBRs in order to efficiently recover the crystals [105,106]. Due to its simplicity and environmental sustainability, MAP precipitation can be also regarded as an elegant method for ammonia disinhibition of HSAD. Nevertheless, literature lacks studies on TAN removal via MAP precipitation in anaerobic digesters treating SS. Xu et al. [107] applied MAP precipitation to remove ammonium and improve sludge stabilization during autothermal aerobic digestion at 6 %TS. The addition of both Mg^{2+} and PO_4^{3-} into the digester was effective in MAP precipitation, as TAN concentration decreased from 2209 to around 1700 mg L^{-1} at the end of the digestion process. As a result, VS degradation kinetics significantly improved after decrease of TAN levels into the digester.

MAP crystallization during HSAD can result in major operational advantages. In conventional WWTPs, MAP has been extensively reported to form massive incrustations on the surfaces of

pumps and centrifuges and cause severe pipe blockage, resulting in additional maintenance and energy costs [108,109]. During HSAD, due to the high viscosity of DSS, MAP is difficult to separate and is mainly retained in the sludge matrix. This can substantially limit operational breakdowns. Additionally, the presence of MAP in the produced ADS enhances its fertilizing potential, being MAP a valuable slow-release source of soil nutrients.

3.2.3.3.Sludge acclimation, bioaugmentation and mixing

Sludge acclimation to high TAN concentrations was shown to play an important role in reducing ammonia inhibition to methanogens during HSAD of SS. Fujishima et al. [110] demonstrated that no inhibition of acetoclastic metabolism occurred at 4.4 g TAN L^{-1} in mesophilic batch reactors with DSS (8.4 %TS) adapted to 3.1 g TAN L^{-1} by cultivation in a continuous digester, although significant inhibition of glucose degradation was observed at TAN concentrations above 0.74 g TAN L^{-1}. In contrast, 3.1 g TAN L^{-1} slightly inhibited hydrogenotrophic methanogenesis during digester operation. Recently, Costamagna et al. [103] and Giordano et al. [31] reported long-term, efficient and balanced HSAD (10.7-12.5 %TS) of SS at TAN concentrations as high as 3.5-4.2 g TAN per kg of ADS in thermophilic digesters at pilot and full scale, respectively. Future studies should further elucidate the potential of sludge acclimation in enhancing the tolerance of methanogens to ammonia during HSAD of SS.

Bioaugmentation has been explored as a possible solution to alleviate ammonia toxicity on AD without interrupting the process and/or changing the feedstock composition. Consortia of SAO bacteria and hydrogenotrophic methanogens are preferably used due their higher ammonia inhibition threshold compared to acetoclastic methanogens [23]. Fotidis [111] observed that

hydrogenotrophic methanogens were the rate limiting organisms of ammonia-tolerant syntrophic consortia in a continuous UASB reactor at TAN concentration in the range of 3-5 g L^{-1}. Bioaugmentation of the fast-growing hydrogenotrophic methanogen *Methanoculleus bourgensis* to a mesophilic CSTR operated at 5 g TAN L^{-1} resulted in a 31.3 % increase of methane production from the digestion of dairy manure. Up to date, bioaugmentation has not been applied yet to overcome ammonia toxicity during HSAD of SS and only a few applications on conventional AD exist in the literature. Lü et al. [112] showed that the addition of the thermophilic bacterium *Coprothermobacter proteolyticus* enhanced the hydrolysis and fermentation of proteins and polysaccharides during thermophilic AD and enhanced methane production by a syntrophic cooperation with methanogenic granular sludge.

Mixing has been reported to effectively ameliorate ammonia inhibition during HSAD as it improves FAN diffusivity and distribution in the digestate. Y. Zhang et al. [38] observed that mesophilic HSAD at 15 %TS in static batch experiments resulted in about 10 times lower methane production yields compared to those (0.19-0.25 L CH_4 g VS^{-1}) previously obtained by Duan et al. [18] in completely-mixed semi-continuous reactors under similar temperature and %TS conditions. Lack of agitation, in fact, may generate microenvironments with FAN levels higher than the average value in the digestate and lead to deterioration of the digestion performance.

3.3.Impact of polyelectrolyte addition on HSAD

Polyelectrolytes are water-soluble polymers with each repeating unit bearing an electrolyte group. Polyelectrolytes are widely used in WWTPs for SS conditioning prior to dewatering at

dosages that typically range between 2.5 and 10 kg per ton TS (as reported in Section 3.1). Polyelectrolyte addition to SS enhances solid-liquid separation according to three main mechanisms: charge neutralization, charge patch formation and polymer bridging [113]. Charge neutralization plays a major role in flocculation with a single polyelectrolyte, while the use of more polyelectrolytes results in enhanced polymer-to-polymer interaction and interparticle bridging [16]. Dose and molar mass of the flocculant as well as process temperature influence dewatering efficiency and must be optimized to reduce the cost of conditioning [114].

Dewatering increases the concentration of polyelectrolytes in SS and may result in detrimental impact on HSAD performance and ADS quality. Indeed, PAM can undergo degradation by a variety of mechanisms, which increase its mobility and the release of acrylamide, a toxic compound. Wang et al. [21] investigated the effect of different cPAM concentrations on WAS in batch and semi-continuous reactors. Increase of cPAM levels from 0 to 12 g kg TS^{-1} lowered methane yield by 38 % and increased the digestion time from 22 to 26 days. About 46 % of the fed cPAM was degraded and only 6.7 % transformed to methane. Acrylamide, acrylic acid and polyacrylic acid are the main degradation metabolites of cPAM and accounted for about 50 % of the degradation products. Polyacrylic acid (32.3 mg L^{-1}) inhibited all digestion steps (solubilization, hydrolysis, acidogenesis and methanogenesis), while detrimental effects of acrylamide and acrylic acid were observed on methanogenesis only.

The impact of polyelectrolyte addition on HSAD was observed to be dependent on the stability of the digestion process. High OLRs can lead to process imbalance and rapid VFA accumulation during the acidogenic step. Litti et al. [115] investigated the influence of the cPAM flocculant Praestol 650 on thermophilic HSAD in batch reactors under optimal and non-optimal ISR ratio.

When the reactors were operated at an optimal ISR of 55/45, the addition of 5-40 g kg TS^{-1} of flocculant to DSS with 7-8 %TS reduced methane production by 50.7 % during the first 13 days of incubation. The inhibitory effect on methane production was attributed to a reduced mass transfer due the formation of thick flocks. The gradual destruction of the floc structure after 13 days resulted in paired methane production between cPAM-treated and untreated batches. At a non-optimal ISR of 40/60, VFA accumulation and subsequent acidification completely inhibited methanogenesis except for the batch reactors with the highest cPAM concentration (40 g kg TS^{-1}), suggesting that bigger flocs played a protective role for microorganisms by generating VFA and TAN gradients.

Thermal-alkaline pretreatment was observed to facilitate cPAM degradation and significantly improve methane production [116]. Pretreating cPAM-flocculated WAS (3.1 %TS) at a temperature of 75 °C and pH of 11 for 17.5 h had a synergistic effect on release, hydrolysis and acidification of OM and destruction of large flocs, enhancing the contact between the released organic compounds and anaerobic bacteria. Floc size significantly decreased after thermal-alkaline pretreatment, while sludge viscosity increased due to the release of extra- and intra-cellular OM to the aqueous phase. This might limit the application of thermal-alkaline pretreatment to DSS, as the elevated viscosity of DSS is a serious issue that severely hampers sludge handling operations such as pumping and mixing.

Future studies should evaluate the impact of different pretreatment and conditioning options on the rheology of DSS in order to optimize polyelectrolyte degradation while allowing sufficient handling properties. Additionally, more biodegradable conditioners should be identified and applied for SS dewatering when upstream HSAD.

3.4.Rheology of DSS

SS can be defined as a non-Newtonian, temperature-dependent suspension of organic and inorganic particles in an aqueous medium. The rheology of non-Newtonian fluids cannot be described by a single value of its viscosity (defined as the ratio between shear stress τ and shear rate σ), being τ not linearly proportional to σ. As a result, an apparent viscosity η (corresponding to a single point of the viscosity function) must be specified [117]. The viscosity of non-Newtonian fluids also depends on their deformation history as a hysteresis loop is generated after removing the shear force. Sludge viscosity decreases as the shear rate increases (*shear-thinning* behavior) due to modification of the sludge structure. This modification is time-dependent and, if the imposed shearing does not exceed the deformation limit of sludge flocs, disappears once the applied force is removed (*thixotrophic* behavior).

Table 5 lists the most used models for describing the rheological behavior of SS. The Herschel-Bulkley model describes sludge as a shear thinning material and provides a valid description of the rheological behavior of SS subjected to various treatment, including AD, dewatering and thermal hydrolysis [118]. A modification of the Herschel-Bulkley model was suggested to better describe the behavior of SS at high shear rates, where the apparent viscosity tends to a limiting value, . Zhang et al. [24] observed that, besides the yield stress τ_γ, DSS exhibited a second critical shear stress τ_c at an intermediate critical shear rate . Therefore, a two-step Herschel-Bulkley model was applied by dividing the equilibrium flow curves into two sections: and .

Table 5 - List of non-Newtonian rheological models used to describe SS and DSS behavior.

Model	Equation	References

Ostwald de Vaele	[15,119,120]
Bingham	[15,121,122]
Herschel-Bulkley	[15,117,118]
Sisko	[123,124]
Casson	[125]
Truncated power-law	[123]
Modified Herschel–Bulkley	[126]
Two-step Herschel–Bulkley , ,	[24]

3.4.1. Impact of solid content on rheological behavior of SS

DSS is a pseudoplastic fluid which exhibits a yield stress and η > 20 Pa s (20000 cP). Yield stress fluids flow only if submitted to a stress above a critical value (τ_γ) and can move from a solid to a liquid state in a reversible way due to the soft interactions existing among the elements composing their structure. The flow characteristics of these fluids are hard to predict as they feature solid and liquid regions difficult to locate [127]. Reducing the yield stress of DSS facilitates management operations such as storage and transportation and is reported to prevent the formation of a sludge crust in the digester [118,128].

SS with a TS content below 2.5-3 % behaves more closely to a Newtonian fluid, meaning that the internal flow resistance is independent of σ and that μ is a constant. Increasing the TS content of the sludge results in higher particle dimensions and interaction forces, which lead to a more pronounced yield stress and viscoelastic behavior [118]. As a result, mixing is severely affected and stagnant regions may form in the digester [129]. Adequate mixing during HSAD of SS can significantly improve biogas yield by enhancing the contact between SS and microorganisms and

alleviating ammonia inhibition, while positive effects in digesters operated at lower TS content are less evident [130]. The mixing ratio of WAS and primary sludge also plays an important role in defining the rheology of DSS. Based on results from Markis et al. [131], the viscosity of raw SS increases by a factor of 5 when the share of primary sludge is decreased from 80 % to 20 %. Füreder et al. [132] observed similar results for DSS, showing that the shear stress of DSS with 8 %TSS increased from 80 to 115 Pa when the share of primary sludge was increased from 40 % to 60 %.

Cao et al. [118] observed a sharp increase of infinite viscosity (defined as the apparent viscosity corresponding to a shear stress of 1000 s^{-1}) and yield stress for both ADS and THS when the TSS content was increased above 7 %. High infinite viscosity and yield stress are undesirable for sludge transport and result in energy-intensive heating and dewatering. Thus, the authors proposed a critical threshold of 7 %TSS beyond which thermal pretreatment and dewatering of the sludge are not cost-effective. Increase in TSS content was also shown to increase the thixotrophic kinetic coefficient, implying that the breakdown and build-up of sludge structure was slower for sludge with higher solid content. Nevertheless, the thixotrophy of ADS and THS showed considerably lower sensitivity to TSS variation compared to untreated SS, which demonstrates that thermal pretreatment is advantageous to mixing during HSAD.

The relationship between shear stress and TS is commonly modeled exponential or power functions [133]. The exponential law tends to overestimate the shear stress at high TS concentrations, whereas using the power function leads to an underestimation of the shear stress values. In a recent study, Füreder et al. [132] showed that the exponential law could successfully describe the TSS-shear stress relationship of both WAS ($R^2 = 0.93$) and ADS ($R^2 = 0.86$).

Increasing TSS from 6 % to 8 % at least doubled the shear stress of raw SS and significantly affected pipe dimensioning and pumping. Nevertheless, under real operating conditions, friction loss calculation based on a simple power law model proved to be sufficiently accurate to describe the rheological behavior of both raw SS and ADS at high TSS.

Effective mixing and pumping of DSS during HSAD can be performed using specific pumps, e.g. PCPs or piston pumps. PCPs transfer the fluid by means of a turning rotor that pushes the sludge forward by generating a sequence of fixed-shape cavities progressing within an elastomeric stator [134]. In order to minimize the wear of the stator due to DSS friction, the rotation rate should be maintained below 130 rpm. An alternative to PCP is represented by high-pressure piston pumps, which feature two pumping cylinders of small diameter and long stroke. Sludge is fed into pump by gravity and the suction created due to the motion of a horizontal piston and a semi-rotary valve. It was reported that a pressure of 4000 kPa was needed to achieve thixotropic breakdown of sludge after a day at rest [134].

Energy consumption for mixing depends on digester size, solid content, type of substrate, feeding time, digester geometry, type of pump/impeller, mixing intensity and mixing strategy. As a result, the specific power consumption for mixing can vary significantly between different AD plants [130]. Although mixing accounts for a major share of electricity consumption in full-scale AD plants, literature lacks evaluation of specific electricity consumption during HSAD of SS. Future research should focus on economic optimization of HSAD plants by improving digester geometry as well as mixing strategy and intensity in order to reduce energy demand for mixing.

3.4.2. Impact of HSAD operating conditions on ADS rheology

AD reduces the strength of the internal structure of sludge particles, resulting in a decreased flow resistance and hysteresis area. By decreasing the TS content, HSAD can considerably improve the flowability of DSS. Zhang et al. [24] showed that all investigated rheological parameters of raw DSS (20 %TS), i.e. yield stress, viscosity, critical shear stress storage modulus (G') and loss modulus (G''), decreased with digestion time. G' decrease followed a linear relationship with VS/TS ratio and TOC and was suggested as a potential controlling parameter for the AD process.

The digestion time plays an important role in determining the rheological behavior of DSS. Füreder et al. [132] observed that digested DSS (6-8 %TSS) with a SRT of 20 days had a higher shear stress and friction loss compared to sludge with a 25-day SRT. Dai et al. [117] observed that ADS from HSAD (16 %TS) with SRT of 30 days resulted in lower shear stress, viscosity and yield stress compared to 20-day SRT sludge under both mesophilic and thermophilic conditions. The positive effects of SRT on sludge rheology can be attributed to the lower TS content of DSS digested with longer SRT. However, temperature conditions and moisture distribution can also affect the rheology of DSS. Despite a lower TS content, mesophilic ADS showed poorer flowability compared to thermophilic ADS, which was attributed to the difference in moisture distribution of the two sludge types. AD was shown to decrease the content of bound and surface water, whereas free and interstitial water content increased. Thermophilic ADS showed a higher content of free and interstitial water compared to the mesophilic one, while the latter showed a higher content of surface water. As a result, thermophilic AD was shown to improve the flowability of DSS more than mesophilic AD and can be considered an advantageous option for the treatment of DSS.

3.5.Biogas contamination during HSAD

AD has been increasingly used for the treatment of industrial wastewaters, resulting in a diversified and case-specific biogas composition. Although CH_4 and CO_2 are the main constituents of biogas, a large variety of trace compounds from household and industry are also present. Trace components include H_2S and VOCs such as siloxanes, terpenes, mercaptans and halogenated compounds, which need to be eliminated to prevent corrosion and excessive wear inside biogas combustion engines. The concentrations of these contaminants during HSAD are considerably higher than those of traditional AD and can severely limit the energetic utilization of biogas. Detrimental effects and typical concentrations of the most common WWTP biogas contaminants are here described in detail, with a focus on their occurrence and implications in HSAD.

3.5.1. Hydrogen sulfide

H_2S is a colorless and poisonous gas with a characteristic smell of rotten eggs even at small concentrations (0.05-500 ppm). During AD, H_2S and other sulfide compounds (dimethyl sulfide, methanethiol and mercaptans) are generated by the anaerobic degradation of sulfur-containing proteins. Intestinal bacteria represent the main source of proteins in SS and, thus, can be also considered the primary source of H_2S in WWTP biogas [135]. Sulfate reduction during thickening and AD also contributes to H_2S generation [25].

Typical H_2S concentrations in biogas range between 50 and 10000 ppm depending on the organic composition of the substrate [136,137]. WWTP biogas usually presents H_2S concentrations well below 1000 ppm and negligible concentrations have been often reported [138]. In WWTP

performing chemical phosphorus precipitation, the addition of iron salts significantly reduces H_2S concentration in biogas by precipitating sulfide as Fe_2S_3 and S^0 [139]. H_2S levels below 1000 ppm enable biogas utilization for energy generation in traditional boilers and internal combustion engines, while much lower concentrations (< 20 ppm) are required for biogas utilization as vehicle fuels and for injection in the natural gas grid [25]. The combustion of H_2S-containing biogas generates SO_2 which, in contact with water vapor and oxygen, produces sulfuric acid (H_2SO_4). Elevated H_2S levels in biogas cause the severe corrosion of piping system, compressors, pressure regulators, gas meters, etc. and the emission of unhealthy hazardous compounds, i.e. SO_2 and H_2SO_4, being responsible for acid rain. As a result, high H_2S levels significantly reduce the economic potential of biogas [140,141]. Additionally, high levels of un-ionized H_2S can be toxic to methanogens. Previous studies reported that 50 % inhibition of methanogenic activity was caused by free H_2S levels between 50 and 270 mg L^{-1} at circumneutral pH [11,142,143]. Koster et al. [144] evidenced that the pH range exerts a significant influence on inhibitory effects of sulfide on the activity of acetoclastic methanogens. Increase in pH from 6.4-6.6 to 7.8-8.0 lowered the inhibitory threshold of free H_2S from 246 to 90 mg S L^{-1}, although the inhibitory levels of total sulfide significantly increased (from 357 to 841 mg S L^{-1}). Besides increasing FAN concentration, increase in pH also reduces the tolerance of methanogens to free H_2S and may lead to a considerable loss of methanogenic activity.

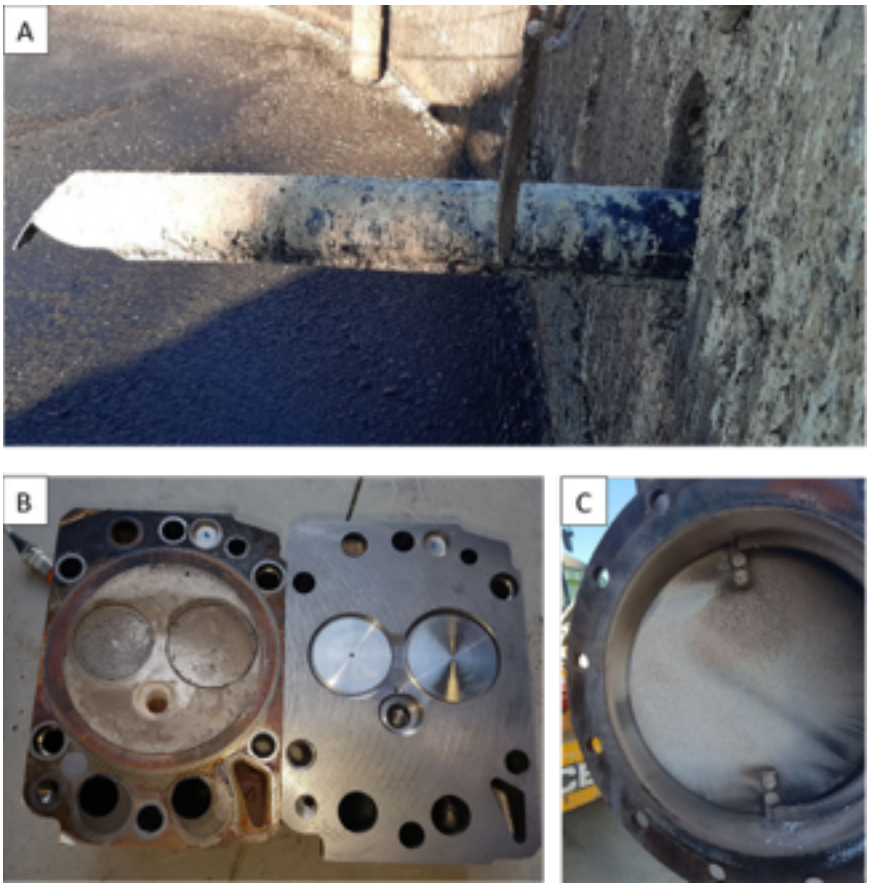
HSAD is expected to generate a higher amount of H_2S in biogas compared to traditional AD due to increased protein concentration in DSS. Nevertheless, Giordano et al. [31] observed that H_2S concentration in the biogas produced by full-scale thermophilic digesters treating DSS (10.7 %TS) did not exceed 2500 ppm. These relatively low H_2S concentrations were attributed to the

high TAN (3.5–3.8 g TAN kg ADS^{-1}) and FAN levels occurring in the digester, which may have inhibited both protein degradation and SRB activity [88,92]. The authors investigated the impact of microaeration at residual oxygen concentrations in the headspace from 0.2 % to 2.0 % (v/v) on H_2S removal from biogas, reaching values below 10 ppm at all tested conditions. By increasing FAN levels (while remaining below inhibitory levels), thermophilic HSAD could limit S^0 deposition on the internal surfaces of the headspace (**Figure 2A**), which is highly advantageous as it limits the interruption of the anaerobic process for cleaning operations. As a result, microaeration can be regarded as an effective and low-cost technique for desulfurization of biogas during HSAD and has been also reported to improve the organic degradation of the sludge [31,145,146].

3.5.2. Siloxanes

Siloxanes are a group of OSC characterized by Si-O bonds in linear (L), cyclic (D) or tetrahedral formation with organic groups (i.e. methyl, ethyl, etc.). Despite their high molecular weight, siloxanes feature a high vapor pressure and low water solubility, resulting in a high Henry's law constant, which indicates that these compounds easily move from water to gas. Siloxanes and other OSC are widely used as emulsifiers, emollient and to produce cosmetics, being inert, safe to use and aroma-free. As a result, part of the siloxanes ends up in WWTPs and adsorb onto EPS of sludge flocs [147]. Additionally, silicon-based compounds are sometimes added to the digester due to their anti-foaming properties and can biodegrade into siloxanes [148].

Most of the siloxanes entering AD volatilize and end up in biogas. Biogas combustion results in siloxane oxidation to microcrystalline silica (SiO_2), a residue similar to sand, which deposits on



gas turbines, valves and pistons (**Figure 2B, C**) causing blockage and abrasion of the mechanical components. Silica also acts as thermal and electrical insulator, causing overheating and failure of spark ignition engines used for electricity production. Additionally, silica deposits are responsible for the deactivation of the oxidation catalysts applied to remove harmful compounds, i.e. hydrocarbons, VOC, CO and NOx, from biogas [149]. The amount of VMS found in WWTP digester biogas can reach 140 mg m^{-3}, which is significantly higher than the recommended limit (15 mg m^{-3}) for safe utilization of most equipment [150].

Figure 2 – (A) Deposits of elemental sulfur on the internal walls of a full-scale thermophilic digester performing HSAD of SS (~11 %TS) under microaerobic conditions. Below, silica

deposits on (B) a dismissed cylinder head compared to a new one and (C) a pneumatic valve installed on the exhaust gas line of the same HSAD plant.

Table 6 reports the concentrations of several siloxanes measured in WWTP biogas at various sites in Europe and USA. Siloxane levels in biogas increase with both temperature and liquid-phase concentration according to Henry's law:

…...(9)

where and are the gas- and aqueous-phase concentrations of the specific siloxane compound, respectively, and is the dimensionless Henry volatility. Siloxane concentrations in HSAD biogas are expected to be much higher than those occurring during conventional AD, as dewatering increases siloxane concentration in SS and thermophilic temperatures are often applied to sanitize ADS and increase biogas production. Therefore, treatment measures are necessary in order to prevent irreversible damages to mechanical components.

Besides volatilization, siloxanes can undergo hydrolysis and adsorption onto sludge. Hydrolysis rate is temperature-dependent as shorter half-lives are observed at higher temperatures [151]. Gatidou et al. [152] used thermodynamic data to predict the contribution of hydrolysis, volatilization and adsorption to the fate of siloxanes during mesophilic and thermophilic AD (2.5 %TSS). Volatilization was observed to be the major mechanism and was enhanced under thermophilic conditions, reaching 99.8 % of siloxane removal from ADS. The contribution of hydrolysis and adsorption was less important, being maximum 8.2 % (for D3) and 3.9 % (for D5), respectively. Thermophilic conditions enhance the hydrolysis rate by reducing the half-lives of siloxane compounds, while siloxane adsorption onto ADS is negatively affected [151,152].

Current technologies for the removal of siloxanes from biogas include adsorption onto activated carbon (being largely applied at full-scale) or solvents (wet scrubbing) [153], chilling and condensation [154], membrane filtration [155] and biodegradation [150]. Sludge pretreatment methods such as pre-aeration [156], pre-digestion [152] and thermal pretreatment with biogas stripping [157] were shown to be effective at lab-scale level for siloxane removal before AD, therefore reducing the need of biogas treatment. However, the effectiveness of these pre-treatment methods for siloxane removal from DSS should be elucidated in future studies.

3.5.3. Volatile organic compounds

VOCs including terpenes, VAHs, mercaptans and halogenated compounds are usually present at trace levels in biogas from AD of SS [158]. Significant concentrations of terpenes, e.g. limonene and cymene, can be found in biogas from co-digestion of SS and bio-waste [159]. Aromatic and chlorinated hydrocarbons, which are used as solvents, foaming agents and propellants in industry, can also occur in SS biogas. Rasi et al. [138] reported toluene concentrations above 10 mg m^{-3} in biogas from a WWTP digester, while benzene concentrations were significantly lower (**Table 6**). Concentrations of halogenated VOCs up to 1.28 ± 0.82 mg m^{-3} were reported in digester biogas analyzed at Yonkers WWTP in New York [160].

Although VOCs have no remarkable detrimental impact on energy production from biogas, they can cause serious damage to industrial installations even at trace concentrations. Hydrocarbons, organosulfur and organochloride contribute to clogging and corrosion of combustion engines [138,159]. Halogenated compounds in biogas can corrode delicate plant components, degrade fuel cells catalysts (e.g. nickel) and reduce the adsorption efficiency of the activated carbon used

for biogas purification [161]. Terpenes were observed to cause deterioration and swelling to rubber materials, which are commonly used in biogas plants for valve sealing and pipe connection. The detrimental effects increase when the rubber is exposed to terpenes in liquid (e.g. condensate) and at high temperatures [162]. HSAD of SS could result in higher levels of

Table 6 - Concentration ranges of trace contaminants in biogas from several WWTPs in Europe and USA.

Compound	Formula/ Abbreviation	Concentration (mg m^{-3})	Reference
Hydrogen sulfide	H_2S	771 – 7580	[163]
Trimethylsilanol	TMS	< 0.02 – 0.09	[154,159,164,165]
Hexamethyldisiloxane	L2	< 0.01 – 0.08	
Octamethyltrisiloxane	L3	0.05 – 0.28	
Decamethyltetrasiloxane	L4	< 0.01 – 1.29	
Dodecamethylpentasiloxane	L5	< 0.01 – 0.8	
Hexamethylcyclotrisiloxane	D3	< 0.01 – 0.286	
Octamethylcyclotetrasiloxane	D4	1.5 – 10.1	
Decamethylcyclopentasiloxane	D5	1.8 – 124	
Dodecamethylcyclohexasiloxane	D6	0.09 – 0.5	
Halogenated compounds		< 0.1 – 1.4	[159]
Benzene	C_6H_6	0.1 – 0.3	[138]
Toluene	C_7H_8	2.8 – 11.8	

biogas contaminants compared to conventional AD, resulting in significant impact on industrial equipment. Knowledge of trace contaminants and their concentrations in biogas during HSAD of SS is therefore of great importance for choosing suitable materials for biogas transport and valorization.

Biogas clean-up technology should be selected based on the target contaminants, regulatory limits and economical investment. GAC has shown high removal performances for several VOCs, including toluene, siloxane D4, isopropanol, ethyl mercaptan both at biogas and saturation concentrations. Additionally, GAC can be regenerated when exhausted by thermal desorption of the adsorbates, although siloxane, ethyl mercaptan and non-volatile organics may severely hamper this process [166]. Mercaptans can also be removed simultaneously with H_2S by alkaline scrubbing and chemical adsorption onto iron sponges. Popular removal systems such as water scrubbing and PSA were shown to be effective for the removal of halogenated compounds [167].

4. Agricultural utilization of high-solid ADS

4.1. HSAD effect on sludge properties

Anaerobic digestion acts positively on the agronomical and hygienic properties of SS. The biological degradation of OM leads to high biological stability, increasing the amendment properties of ADS [168,169]. Recent findings revealed that HSAD led to the ADS acquiring high biological stability (measured by both anaerobic and aerobic tests) (**Table 7**), which was higher than that of compost. Additionally, HSAD provided a high maturity rank, i.e. absence of phytotoxicity, with benefit for plant production [168]. This status resulted in reduced odor

emission for ADS during distribution on agricultural land [170]. Moreover, the concentrations of organic pollutants, i.e. total hydrocarbons, toluene, pesticides, AOX, LAS, DEHP, NPE, PAH,

Table 7 - Chemical and biological characteristics of high-solid ADS.

	Odors	pH	VS	TOC	TKN	N-NH_3	OD_{20}[c]	ABP[d]	P	K	Fe	Ca	Mg
	OU $m^{-2}h^{-1}$		g $kgTS^{-1}$	g $kgTS^{-1}$	g $kgTS^{-1}$	g $kgTS^{-1}$	mg O_2 gTS^{-1} 20 h^{-1}	NL $kgTS^{-1}$	g $kgTS^{-1}$	g $kgTS^{-1}$	g $kgTS^{-1}$	g $kgTS^{-1}$	g $kgTS^{-1}$
HSAD[a]	3706	8.3 ± 0.2	555 ± 0	289 ± 12	52.3 ± 2.3	20.8 ± 0.2	33 ± 1	134 ± 4	22.4 ± 0.2	3.2 ± 0.0	45.6 ± 0.1	25.8 ± 0.0	5.6 ± 0.0
HSAD[b]	3740 ± 846	8.6 ± 0.3	606 ± 0	320 ± 42	77 ± 7	45 ± 3	22 ± 6	35 ± 4	30 ± 4	6.3 ± 1.4	25.9 ± 3.9	55.5 ± 5.5	4.6 ± 0.0

	As	Cd	Cr	Cr^{6+}	Cr^{3+}	Hg	Pb	Ni	Cu	Zn	Mn	Mo
	mg $kgTS^{-1}$											
HSAD[a]	5.6 ± 0.4	2.4 ± 0.1	197 ± 1	0.05	196.9 ± 0.6	1.44 ± 0.01	128 ± 1	107 ± 2	411 ± 3	852 ± 8	n.d.	n.d.
HSAD[b]	8.1 ± 2.6	0.9 ± 0.5	78 ± 25	< 0.5	n.d.	< 1.3	58 ± 11	52 ± 10	376 ± 70	960 ± 80	451 ± 30	9430 ± 0

	Total hydrocarbon C10-C40	Nitrogen organic solvent	Aromatic organic solvent - toluene	Chlorate organic solvent	Surfactant	Chlorate pesticide	Phosphate pesticide	AOX	LAS	DEHP	NPE	PAH	PCB	PCDD/F+PCB-DL
	mg $kgTS^{-1}$	mg L^{-1}	mg $kgTS^{-1}$	mg L^{-1}	mg L^{-1}	mg L^{-1}	mg L^{-1}			mg $kgTS^{-1}$				ng TE $kgTS^{-1}$
HSAD[a]	1.95 (mg L^{-1})	< 0.01	0.002	0.003	3.3	< 0.01	< 0.01	0.03	131	18.14	< 0.67	< 0.07	0.46	2.95
HSAD[b]	572 ± 4[e]	n.d.	2.81	n.d.	n.d.	n.d.	n.d.	0.4	n.d.	7.24	< 7.5	n.d.	< 0.1	10.1 ± 2.5

n.d. = not detected.
[a]HSAD pilot scale from Scaglia et al. [168].
[b]HSAD full scale plant data from Zilio et al. [170].
[c]OD_{20}: biological stability measured by respirometric test.
[d]ABP: biological stability measured by anaerobic test, i.e. potential biogas production.

[e]C10-C40.

PCB, PCDD, etc., detected in high-solid ADS are very modest (**Table 7**) and well below the regulatory limits for its agricultural utilization [3].

By degrading OM, AD increases the concentration of mineral N and P forms in the ADS at the expense of the organic ones, increasing nutrient availability to plants [169]. Previous investigation indicated that high-solid ADS has better fertilizer properties compared to untreated SS, compost and agriculture digestate as well as amendment ability similar to those of compost [168]. These results were due to the high mineral content of N and P and biological stability of OM (**Table 7**).

HSAD has been reported, also, having a strong effect on pathogen reduction in SS. *Salmonella* spp., *Escherichia coli*, fecal streptococci, *Enterobacteriaceae*, fecal coliforms and helminth eggs were reduced to below the detection level, and even the population of *Clostridium perfrigens* was strongly reduced [171]. This result was achieved because of the bacteriostatic activity made by high pH, high ammonia due to protein degradation and high biological stability acquired with the biological process [171,172]. In addition, thermophilic temperatures guarantee higher sanitation performances, outperforming those obtained under mesophilic and psychrophilic conditions [171,173].

4.2.Effect of high-solid ADS application on soil

SS and ADS can have different physical composition ranging from a liquid form with < 5 %TS to dewatered material to completely dried and pelletized material (> 90 %TS) [174]. TS content affect distribution modality in field, i.e. spreading, injection and incorporation into soil [174].

Generally, spreading and injection are devoted to pumpable material, contrarily to incorporation used for solid/palatable products.

HSAD allows getting a pumpable final product having high TS content, i.e. > 10 % ww, which has some advantage for agronomical purpose. First, the high TS content allows high OM and nutrient concentration when referred to the wet matter, i.e. less product needs to be spread in the land. In addition, pumpable products can be injected directly into soil by applying a VRT control, further reducing the problems connected with both odor and aerosol annoyance and with ammonia emission. It has been demonstrated by a full-scale approach that injecting high-solid ADS results in NH_3, N_2O, CH_4 and odor emissions similar to those of synthetic mineral fertilizers [170]. However, injection operations should be carefully monitored and optimized since the high viscosity of high-solid ADS may lead to its uneven distribution on the agricultural field due to different quantity injected by the anchors.

High-solid ADS, being rich in stabilized OM, plays an important role in affecting soil physical properties above all when dosed to heavy-textured and poorly structured soils, i.e. amendment action. The high presence of electric charged colloidal organic molecules affects the soil properties by forming stable aggregates with soil mineral particles, decreasing bulk density and increasing soil porosity. Water retention, also, is affected because of the ability of charged OM to form hydrogen bonds. All these have been well documented for ADS application on soil [174,175].

High-solid ADS contains macro- (N, P, K), meso- (Ca, Mg, S, Fe) and micro- (e.g. Mn, Zn, Cu, Mo, Co) nutrients (**Table 7**) that exert a fertilizer action on soil, such as widely reported in the

literature for SS [175]. In general, these elements are present under medium-readily availability forms because of chemical-biological modification occurring during anaerobic digestion [169] and high presence of OM protecting macro- (P), meso- (Ca and Mg) and micro-elements from chemical precipitation/insolubilization. Therefore, high-solid ADS has been proposed as a substitute to synthetic fertilizer for crop production. Full-scale trials confirmed the fertilizing potential of high-solid ADS, i.e. after two years of experiment there was no difference in crop (corn) production for the soil fertilized with digestate and that with chemical fertilizer [170].

5. Guidelines for cost-benefit analysis of HSAD plant treating SS

Cost-benefit analysis for a HSAD plant treating SS is very case-specific as it strongly relies on the following criteria:

1) *Disposal costs for agricultural recovery of SS.* The primary driver behind the investment for a centralized HSAD plant is the recycling of organic waste to organic fertilizers. The increasing utilization of SS-derived fertilizers as a replacement of synthetic mineral fertilizers would lead to substantial economic savings for farmers while promoting circular economy. This marks a fundamental difference with WWTP digesters, for which several disposal options are potentially applicable depending on ADS quality and local conditions. The cost for disposal of SS may vary significantly depending on the country and disposal method. The attractiveness of agricultural recovery of SS depends on the competitiveness of alternative disposal methods existing in the region, e.g. incineration, landfilling and composting. However, the agricultural reuse of SS should be prioritized as recommended in the Circular Economy Action Plan by the European Commission [3].

2) *HSAD plant position.* One of the main advantages of HSAD of SS is the possibility to use ADS for agronomic purposes, therefore reducing the amount of SS disposed via other methods. In this context, the geographical position of the plant is crucial, as the abundance of arable agricultural land increases the share of SS processed for agricultural recovery. From an economical point of view, investment into HSAD of SS is strongly influenced by the costs of land application, transportation and injection of the digestate, which increase considerably with transportation distance [176].
3) *Incentives for energy production.* The existence of subsides to produce green energy (electrical energy and/or biomethane) could significantly impact the cost-benefit analysis of the HSAD plant.
4) *Type of cultivation.* The amount of digestate required for agricultural cultivation also depends on the type of culture, which influences the demand for fertilizing products.

Table 8 lists revenues and costs of a HSAD plant recycling SS for agronomic application. The main revenue of the plant is represented by the gate-fee for substrate supply. SS availability is commonly not an issue. In Europe, the implementation of the urban wastewater treatment Directive D91/271/EC and Commission Directive 98/15/EC in the past years has led to a strong rise in sludge generation within EU27 [177]. Nevertheless, public acceptance of SS-derived fertilizers still represents a challenge. Therefore, much attention and significant investment should be dedicated to ADS hygenization as well as to the abatement of odors and aerosol from the plant and during ADS distribution into farmland, which can be achieved by closing DSS unloading facilities, injecting ADS directly into the soil and ensuring its biological stability.

Another revenue of the HSAD process is represented by the production of electrical energy, which is primarily used on site and the excess fed to the national grid, or biomethane from biogas upgrading. A third source of revenue can derive from sales of NR products, such as the ammonium sulfate produced via ammonia stripping if needed to sustain the HSAD process.

Costs related to HSAD plant operation concern personnel, overhead expenses, maintenance, financing, purchase of consumables and handling, transportation and application of ADS (**Table 8**). The HSAD process requires higher investment and operational costs compared to conventional AD, as specific equipment for pumping, mixing and heat exchange is needed, being more expensive than that commonly used for WWTP digesters.

Table 8 – Revenues and expenses summary for a HSAD plant for agricultural valorization of SS.

Revenues	**Expenses**
• Gate-fee for SS supply • Sale of surplus electrical energy or biomethane • Sale of NR products	• Consumables (chemicals, spare parts) • ADS handling (storage, transportation and application) • Operations (personnel, overhead, maintenance) • Finance (amortization and interest)

6. Conclusions

HSAD is a promising bioprocess for both energy and material recovery from SS, as it results in an improved energy balance compared to conventional AD and in the production of ADS with a

significant agronomic value in terms of humified OM and nutrient concentrations. Additionally, HSAD represents an opportunity to rationalize the wastewater treatment system by centralizing treatment and valorization of DSS and leaving the sole wastewater treatment to small and decentralized WWTPs. In order to efficiently operate the HSAD process, appropriate technologies should be applied for DSS mixing, transportation and pretreatment, process control, biogas purification and ADS distribution on agricultural soil. Future research should primarily focus on improving HSAD efficiency by tailoring pretreatments to chemical and rheological properties of DSS and further enhancing remedial actions to process inhibition. Also, the fate of emerging contaminants during HSAD of SS and their impact on the process performance should be investigated. Biogas pretreatment methods should also be enhanced in order to extend the service life of industrial installations and reduce maintenance costs.

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