

Review

Review of the Integration of Drying and Thermal Treatment Processes for Energy Efficient Reduction of Contaminants and Beneficial Reuse of Wastewater Treatment Plant Biosolids

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Abstract: Biosolids have been traditionally used as a beneficial resource in the agricultural industry. However, contaminants of emerging concern (CECs) threaten their reuse due to concerns of toxicity, bioaccumulation, and increased regulations on acceptable CEC concentrations in biosolids. The thermal treatment of biosolids has the potential to destroy/mineralize these contaminants as well as transform the biosolids into valuable biochar. However, the thermal processing of biosolids is highly energy intensive due to the energy costs associated with drying biosolids to the required moisture content for thermal processing. This article performs a brief review of the drying of biosolids from a physical and theoretical viewpoint. It also provides an overview of pyrolysis and gasification. It explains the impact that moisture can have on both the degradation of CECs and the products that can be obtained through the thermal treatment of biosolids. Additionally, model-based, lab-based, and pilot-scale examples of integrated drying and thermal treatment processes are reviewed. Key challenges, such as the need for co-pyrolysis and co-gasification, as well as the impact of biosolids composition on energetic viability, are identified.

Keywords: biosolids; sewage sludge; drying; thermal treatment; pyrolysis; gasification; energy integration



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1. Introduction

Accelerated industrialization, urban development, and population growth have led to a substantial increase in the use of natural resources and the production of waste [1]. Consequently, there has been an increased focus on creating more sustainable circular economies (CE) across industrial sectors. Wastewater treatment plants (WWTPs) have become the focal point for such research due to the rich potential for resource and energy recovery from these facilities [2], especially biosolids (BS).

BS are usually a complex heterogeneous mixture and can range in moisture content (MC) from 20 to 90% (wet basis) [3]. Factors such as process generation, age, and environmental conditions can affect BS composition [4]. Organic matter constitutes 50–70% of dry BS composition; fatty acids, steroids; amino acids; nucleic acids, lipids, and polysaccharides [5,6]. Essential macronutrients that can be found in BS include phosphorus, nitrogen, potassium, and sulfur [2,7]. Nitrogen, phosphorus, and potassium are typically considered the three most important elements for plant growth, with most fertilizers being specified by their NPK (nitrogen, phosphorus, potassium) ratio. Phosphorus is considered a critical resource due to its necessity in plant development and having no viable substitute. With fertilizer production being a major global challenge for addressing climate change, the reuse of BS forms an important step towards sustainability.

Traditionally, BS have been dealt with by landfilling, incineration, or usage in agricultural applications [8,9]. Landfilling and incineration are costly, emission-intensive processes that do not lead to the beneficial reuse of valuable constituents in BS [1]. Agricultural use of BS enables the recycling of key micro and macronutrients, making BS a valuable resource as a fertilizer and/or soil amendment. This reuse reduces the need for synthetic fertilizers

and helps to close the nutrient loop. Hence, the application of BS to agricultural, forestry, and land reclamation projects has internationally been recognized as the most sustainable option for management [1,10–12]. However, WWTPs treat water from various sources, including homes, industrial factories, farms, hospitals, and stormwater run-off, resulting in various contaminants making their way into the final treated effluent and BS, compromising their safe reuse [11,13].

Contaminants of emerging concern (CEC) include both new and emerging pollutants and existing chemicals with only recently recognized health and environmental impacts [14]. Known CECs, among thousands of others, include products such as personal care products (PCP), steroids/hormones, perfluorochemicals (PFCs), and microplastics, all of which have been found in BS [1,8,10,15–18]. Water authorities, researchers, and industry experts are increasingly becoming aware of the persistence and threat that these compounds pose in the environment [13,19]. Furthermore, studies have shown plants/crops uptake CECs, and leaching of such contaminants leads to polluted soils [9,16,20,21]. This can induce plant stress symptoms and ultimately lower crop yields [22,23]. Hence CECs have become a global concern, which has resulted in legislation restricting the safe disposal or sustainable use of BS [1,10,21,24], and the presence of CECs is making land application of BS increasingly infeasible. Some municipalities and countries, such as Switzerland, have banned the use of BS on agricultural land due to concerns about CECs [25].

Furthermore, while legal limits exist for known/traditional contaminants, such as N and P, which are subject to locally relevant government regulation dependent on catchment and treatment plant specifics (see, for example [26]), and heavy metals (e.g., arsenic 20 mg/kg dry [26]), many of the CECs have no legally defined concentration threshold; hence, they are seldom monitored in wastewater facilities. There is considerable literature regarding traditional contaminants in biosolids, their impacts, and the required treatment processes to remove them; hence, they are not covered in this work. Traditional WWTPs are mostly ineffective at removing CECs [19,27]; therefore, novel treatment methods are required to ensure BS can continue to be beneficially reused. Currently, thermal treatments such as pyrolysis and gasification are the preferred technologies for removing/destroying CECs from solid/sludge matrices. The thermal destruction of CECs relies on thermolysis; therefore, it is crucial to maintain the contaminants at destruction/mineralization temperatures for sufficient residence time for complete thermolysis. Such processes have the added benefit of transforming BS into carbon-rich biochar, which shares many of the same nutrient benefits of BS with the added benefit of sequestering carbon [28]. However, the feed moisture content for these treatment technologies has been recommended to be 15% (wet basis) or below to be economically viable [15,29]. Wet BS coming out of a WWTP may range from 85 to 99% MC, emphasizing the importance of critical BS preparation steps: dewatering and drying.

Many WWTPs already include a mechanical dewatering process, which physically removes moisture from the solids. Dewatering processes include centrifugal, belt filter presses, thickeners, and plate filters [30]. Some plants may already include a drying process as part of the WWTP due to the benefit of reducing the mass of the BS, resulting in significantly cheaper handling/transport costs. Moreover, for BS to be safe for land use, heat treatment (whether from drying or composting) is required to kill/destroy pathogens. Treatment specifications will vary for each region; as a local example, this information is detailed in the QLD end-of-waste code [26]. For a European context, readers are referred to Collivignarelli et al. (2019) work [31]. Common dryer types include direct dryers (flash dryers, spray dryers, rotary dryers) and indirect dryers (steam dryers, hollow-flight dryers, tray dryers). Other, more novel drying technologies include microwave drying, fry drying, bio-drying (similar to composting), and solar drying [32,33].

To ensure viability, new technical solutions and processes aimed at the thermal treatment of BS to facilitate reuse should be optimized for energy efficiency [34]. This is especially important because energy costs associated with thermal treatment are not comparable to the energy costs associated with drying [35]. Unless drying is carefully considered

at the process development stage, there is potential that thermal treatment of biosolids for contaminant destruction will be uneconomical and result in a financial burden for WWTP owners. Therefore, because of the coupling between drying and thermal treatment, it is critical that drying technologies are integrated with thermal treatment to effectively utilize any waste-heat arising from thermal processing. Despite such importance and extensive coverage of both drying and pyrolysis/gasification of BS in the literature, there is a lack of papers or reviews dedicated to the integration of these processes. This review paper addresses this gap by first outlining BS drying theory along with biosolid drying characteristics such as cracking and shrinking, which have implications for the practical processing of BS. The review seeks to highlight the variability in both BS composition and BS drying behavior, which influences the efficiency of integrated destruction processes. Next, a brief review of modeling BS drying is conducted, which highlights the challenges in developing accurate and robust models suitable for modeling drying and thermal treatment. Pyrolysis and gasification processes are briefly described, and the fate of CECs during thermal destruction is outlined. Emphasis is placed on describing the influence of moisture content on the destruction mechanisms, which seeks to reinforce the importance of understanding simultaneous drying and thermal processing. Finally, to give a practical perspective, a review of the integration of drying and thermal treatment is described. This is intended to provide examples of process integration that will assist both researchers and practitioners in developing integrated sustainable BS treatment systems. Note that biosolids and sewage sludge are frequently used interchangeably in the literature. This review hereon refers to all such sources as biosolids.

2. Drying of Biosolids

Solid drying is typically represented using a drying rate curve. Usually, curves will show one or two trends, depending on the type of moisture being removed (bound and unbound): constant rate drying and/or falling rate drying. A brief initial adjustment or heating period may also be observed. Constant rate drying refers to the removal of unbound moisture from the material surface, which can be represented by convective mass transfer and partial pressure driving forces. The concentration of water at the surface (and hence the partial pressure) does not change until all the moisture is removed; therefore, the rate does not change. Removal of bound/internal water shows a falling rate as the average moisture concentration is reduced; hence, the concentration gradient with the surrounding gas is also reduced, lowering the overall transfer rate. Multiple falling rates may be observed depending on material properties such as porosity. A typical drying curve is shown in Figure 1.

The most critical information for drying equipment is the time needed to remove the desired moisture from the material. First principles modeling can answer this by solving simultaneous heat, mass, and momentum transfer equations. However, extensive characterization of the material, the environment, and material changes during drying are needed [36,37]. Variables known to affect the drying rate of organic materials include temperature, gas velocity, relative humidity, material thickness (i.e., particle size), and internal material properties [38,39]. The dominant mechanisms for heat transfer in BS dryers are usually convective or conductive, although there is increasing research regarding radiative dryers. Table 1 presents the advantages and disadvantages of the main BS dryer types, along with specific energy consumption and expected drying rates. Due to the highly variable nature of sewage sludge, each WWTP's BS may have different material properties, resulting in different drying behavior [40].

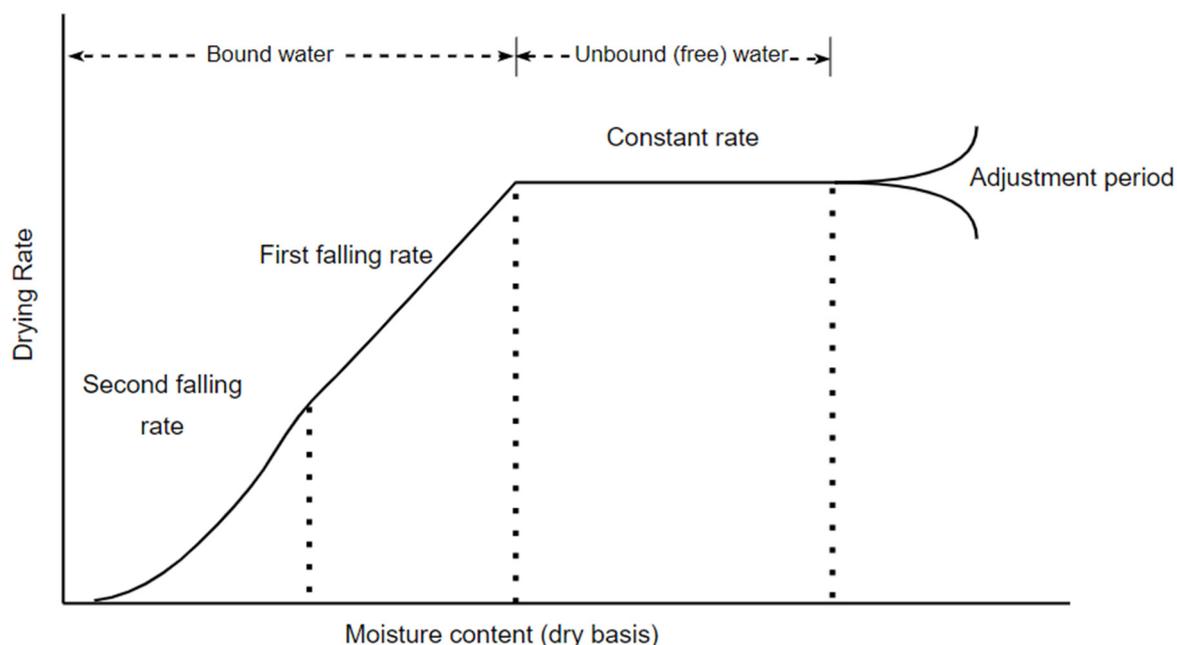


Figure 1. Typical wet solid drying curve.

Table 1. Energy consumption and drying rates of biosolids dryer technologies. Adapted from [41].

Drying Type	Specific Energy Consumption $\left[\frac{\text{kWh}}{\text{ton evaporated water}} \right]$	Specific Drying Rate $\left[\frac{\text{kg}}{\text{m}^2 \cdot \text{h}} \right]$	Advantages	Disadvantages
Convective	700–1400	0.2–30	<ul style="list-style-type: none"> • Easy design manipulation 	<ul style="list-style-type: none"> • Long drying time • Bad odors
Conductive	800–900	7–35	<ul style="list-style-type: none"> • No pollution of the heat medium • Steam and odor confinement 	<ul style="list-style-type: none"> • Long drying time • The sticky phase may alter the performance
Solar	30–200	N/A *	<ul style="list-style-type: none"> • Free solar energy 	<ul style="list-style-type: none"> • Depends on climate conditions • Lots of surface area needed

* Solar drying is too variable to have a consistent range. Background color included for readability.

During drying, BS exhibits various behaviors which can either inhibit or improve the process. Commonly observed behaviors include shrinkage, skin-layer formation, and significant formation of pores/cracks, as shown in Figure 2 [42–45]. Shrinkage and skin-layer formation retard drying, as shrinkage reduces the available surface area for heating and mass transfer, and skin-layer formation introduces a new resistance to water/mass transfer [45]. Cracks, on the other hand, have been shown to increase the drying rate, Due to increased surface area and reduced diffusion pathways [46]. Additionally, as BS dry, they may pass through a “sticky” or “plastic” phase. During this phase, the BS adheres to the surfaces of drying equipment, decreasing mixability and heat transfer, as well as increasing maintenance costs [33]. Properties/composition strongly influence when this phase occurs, as well as the other morphological changes [41]. For example, Leonard et al. (2005) convectively dried two different sludges (with and without pre-treatment to reduce BS nutrient levels. The high nutrient BS exhibited a long constant drying period, suggesting external mass transfer limitations. Whereas the low nutrient BS were characterized by a long decreasing (falling) drying rate, indicating diffusion limitations from the onset of drying. The sludge with low nutrient levels always dried slower. Therefore, it is highly

recommended that individual mapping of each WWTPs BS drying is conducted [47]. A full review of BS drying is beyond the scope of this paper. For greater detail on BS drying, Bennamoun et al. (2013) provide a thorough review of drying BS, including explanations of dryer systems, sludge behavior during drying (cracking, shrinkage), and further discussion of the sticky/plastic phase [41].

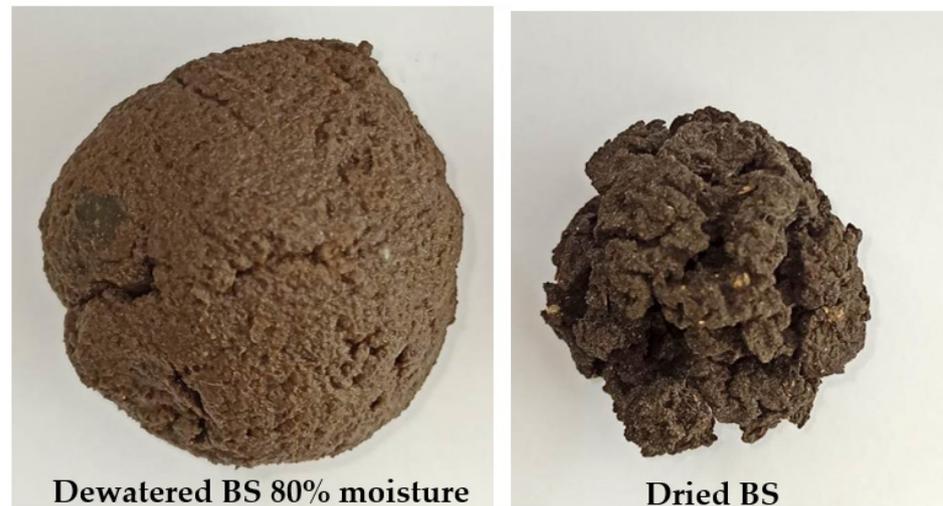


Figure 2. Anaerobically digested biosolid morphological changes (shrinkage, cracking, skin layer formation) during drying. Convective drying at 138 °C and drying gas velocity of 2 m/s.

3. Modelling Moisture Removal from Biosolids

The modeling of drying generally uses one of two models, mechanistic or analytical [36]. The mechanistic approach seeks to describe each mechanism involved in water removal using physically sound relations. Thus, a mechanistic model would include simultaneous heat and mass transfer equations and a good understanding of material properties and boundary conditions. Analytical methods (which can be empirical, semi-empirical, or theoretical) are developed by solving diffusion equations or equation fitting. Diffusion models are the most commonly used to represent solids drying, although they are only applicable for solids under predominately falling rate drying periods [45]. Despite being widely used, empirical models cannot predict how changes in material properties or operational parameters will affect drying time, as the parameters used in these models lack physical significance. Thus, they are only valid under the specific conditions they were developed under. Many researchers base their drying models on the equations of Fick and Fourier, which represent transient mass and heat transfer, respectively [42,43,48,49]. However, correction factors (relating to the impacts of shrinkage and/or cracking) are often needed to produce satisfactory results.

The authors could only identify one review paper concerning BS drying models, focusing only on thin-layer drying models [50]. Hence a brief review is presented here of three studies that provide a general overview of typical work conducted in this area. Table 2 summarizes the experimental conditions and modeling approaches for a selection of the reviewed papers.

Table 2. Summary of drying conditions and models developed.

Drying Apparatus	Principal Heat Transfer	Gas Velocity [m/s]	Drying Temperatures [°C]	Initial Moisture Content	Sample Dimension(s)	Measured Variables	Model(s) Developed	Reference
1-9 Drying tunnel	Convective	0.43, 0.65	80, 90, 100, 112	72.5% wb	0.1 × 0.1 × 0.005 m, 0.1 × 0.1 × 0.009 m	Mass	Fickian, Modified Quasi-stationary, Two-period	[42]
Drying Tunnel	Convective	1.58–1.82	122–158	3 kg/kg dm 2.6 kg/kg dm	12 mm cylinders	Mass	Fickian	[43]
Drying tunnel	Convective	1, 2, 3	80, 90, 140, 200	86, 88, 6% wb	15 mm cylinder	Mass, surface temperature	Empirical	[44]
Drying tunnel	Convective	2.4–5.5	31–64	2.4–4.2 kg/kg dm	2.5–3 cm spheres, cylindrical tablet 1 cm × 6.6 cm	Mass, internal temperature	Fickian	[45]
Drying Tunnel	Convective	1.00	83.7	80–82% wb	26 mm diameter, 10 mm height sludge cylinders	Mass	No drying model developed, cracking model developed	[46]
Satorious moisture analyzer	Radiative	N/A	80, 120, 160	80% wb	2–20 mm spheres	Mass	Fickian	[49]

Background color included for readability.

Zhao et al. (2020) developed a model based on Fick's 2nd law of diffusion after drying spherical sludge particles of various sizes in a rapid moisture analyzer [49]. The principal heat transfer for these experiments was radiative, an unlikely primary heat transfer mode for typical industrial drying apparatuses, which are predominately convective and conductive. The model was developed under the following assumptions: The moisture content inside the particle is uniform; there is no temperature gradient in the particle. To capture the effects of morphological changes (shrinkage and cracking), the effective diffusion coefficient was varied as a function of time. The final model fitted the data well; however, due to its highly empirical nature, it is unlikely to be able to be adapted to any new conditions other than those used to develop the model.

Reyes et al. (2004) dried sludge in open metal boxes in a convective drying tunnel [42]. Considerable shrinking, cracking, and skin layer formation were observed. A constant rate drying period was identified, followed by two falling rate periods. Three models were developed, one based on Fick's 2nd law, a modified quasi-stationary method, and a two-period model. All the models focused on obtaining an average moisture content and did not consider internal temperature or internal moisture distributions. The Fickian and quasi-stationary models were able to sufficiently describe the drying kinetics in the falling rate period, with deviations between the experimental values being attributed to morphological changes during drying, which were not considered in either model. The two-period model was able to simulate the entire drying process (constant and falling rate periods) with a very strong fit. This model was then generalized such that it may be possible to predict drying kinetics at other temperatures and air velocities, although this was not verified.

One of the few papers that investigated internal temperature distribution during drying was by Font et al. (2011) [45]. BS spheres and cylinders from two different WWTPs were used. Internal temperature measurements were made by inserting a thin thermocouple into the spheres. Both sludges exhibited shrinkage, skin layer formation, and cracking; however, one sludge had a much stronger skin layer formation. Additionally, a particle was sliced in half before drying had completed, which showed that the particle could be classified into three parts, a wet inner core, an intermediate section, and a dry external solid.

Font et al. (2011) developed a numerical model by dividing the spherical particles into 20 volume elements of equal dry solid mass. The mechanistic heat and mass diffusion equations were then discretized according to finite differences. Two correction factors, expressed as functions of moisture content, were added to the surface mass transfer boundary conditions.

One correction factor aimed to capture the effect of the skin layer (which retards drying). This factor varies from 1 at the beginning of drying to nearly zero when the moisture is close to the equilibrium moisture. Hence, as drying progresses, there is an added resistance to mass transfer. However, the correction factor varied significantly depending on the sludge (0.3 for one and 1.3 for the other). The second correction factor aimed to capture the effect of shrinkage. Shrinkage was related to the average moisture content of the particle, with the correction factor varying from 1 at the beginning of drying to 0.7 by the end of it. Satisfactory results between the model and experimental data were produced after optimization of the correction factors. However, the correction factors were considerably different for the two sludges, highlighting the importance of physical changes that occur in BS as they dry and the need to characterize any BS being considered for thermal processing.

From the reviewed papers, it is evident that sludge drying behavior is highly dependent on sludge origin. Therefore, to develop a model for the optimization of drying equipment, there is likely a need to obtain experimental data on each WWTP's BS to capture their specific skin/shrinkage/mass transfer resistance parameters. Moreover, due to potentially significant morphological changes during drying, modeling is necessarily complex, limiting the potential application of empirical models and supporting the use of mechanistic modeling. However, though modeling will require experimentation, a few simple experiments can significantly enhance the models developed, making them fit for purpose. Such a validated model will facilitate the cost-effective investigation of other operational variables and their impacts without having to perform additional experiments. Furthermore, the use of a model-based design approach will enable the optimization of drying and thermal processing equipment. As will be discussed in the following section, accounting for internal moisture and temperature profiles is important when integrating drying and thermal treatment processes. However, such models are limited in the literature, with internal conditions frequently ignored. Similarly, studies investigating the internal temperature distribution of BS are limited.

4. Thermal Treatment of Biosolids

Thermal processes use heat energy to transform materials, with combustion being the most basic heat treatment and more complex thermal treatments being pyrolysis, gasification, and hydrothermal liquefaction. Combustion does not allow for the beneficial agricultural reuse of BS and releases unsustainable CO₂ emissions; therefore, it will not be reviewed. Likewise, hydrothermal processes do not require drying and are not reviewed. The most fundamental difference between pyrolysis and gasification is the environment they are performed in. Pyrolysis is performed in an inert atmosphere, whereas gasification uses a sub-stoichiometric amount of oxygen. Both pyrolysis and gasification transform the BS into biochar, although pyrolysis generally produces more biochar than gasification. Biochar can be used as a soil amendment to simultaneously improve a broad range of soil properties, increase agricultural yields, and contribute to climate change mitigation via carbon sequestration [51,52]. Furthermore, pyrolysis and gasification reduce BS volume and mass by up to 70%, allowing for cheaper handling and lower transport costs. For a more comprehensive analysis in this area, Gao et al. (2020) reviewed processes involved in the thermochemical conversion of sewage sludge/BS, focusing on the use of blended feeds (i.e., co-pyrolysis) as well as product distributions and product composition and their relationship to thermal treatment process conditions [15].

4.1. Pyrolysis

Pyrolysis is the thermal decomposition of material in an inert atmosphere (oxygen-free) at elevated temperatures. It may be performed in either fixed or fluidized beds. Fluidized beds allow better gas-solid contact, allowing for faster and more uniform heat distribution [53]. Temperatures range from 350 to 800 °C, with the inert environment usually being N₂. Three products result from the pyrolysis of carbonaceous materials such as BS:

biochar, a residual solid phase; py-oil, a liquid phase that condenses upon cooling; py-gas, a permanent gas phase. Biochar can be used as a soil conditioner, with reduced leaching potential (compared to unprocessed BS), or as an adsorbent or catalyst [54]. It is also considered a carbon sequestration technology as the converted carbon does not readily degrade, with residence times estimated to be over 1000 years [55]. Furthermore, biochar has been shown to be a carbon sink, able to adsorb CO₂ from the atmosphere [56]. Py-gas contains hydrogen, carbon monoxide, methane, carbon dioxide, and other hydrocarbon gases [35]. Py-oil is a mixture of hydrocarbons, oxygenated and nitrogenated compounds [54]. Both the py-gas and py-oil can be burned in industrial boilers to provide heat/energy to the pyrolysis process, with surplus heat provided for drying. Py-oil may also be utilized as fuel after upgrading [57]. The ratio and composition of these products depend on the mode of pyrolysis used. For greater detail regarding pyrolysis and the different modes (fast, slow, flash, etc.), Elkhalfia et al. (2022) recent work reviewed the pyrolysis of BS to biochar [58].

Pyrolysis can be broken into two or three main stages, depending on initial moisture content: dehydration, devolatilization, and thermal degradation. Ideal moisture content for economic viability varies, and specific analysis based on BS type and treatment conditions is necessary. Generally, pyrolysis requires a feed moisture content of under <15% [59–61]; some studies state that pyrolysis can be energy neutral/positive at moisture contents up to 50% [35,62]. Moisture has also been shown to be beneficial to the pyrolysis process with increased production of hydrogen-rich py-gas. The presence of moisture also promotes the cracking and reforming reactions of aromatic compounds [63].

4.2. Gasification

Gasification is the partial oxidation of carbonaceous materials in an elevated temperature environment. The process is tolerant of diverse feedstocks. Temperatures are typically higher than pyrolysis and range from 500 to 1200 °C at atmospheric pressures. A sub-stoichiometric amount of oxygen is provided, usually 65% less than that required for complete combustion [64]. Products of gasification include a gaseous fuel composed of largely N₂, H₂O, CO₂, CO, H₂, CH₄, and other low-weight hydrocarbons; tars, ash, and biochar [65]. Similar to pyrolysis, these products can be burnt in industrial boilers to generate heat/electricity, and biochar can be used for agricultural/soil amendment purposes.

The principal stages during gasification include drying, pyrolysis, oxidation (gas-solid reactions), and reduction (gas-phase reactions). A distinct advantage of gasification is that the initial moisture content can be higher than other thermo-chemical processes, with MC potentially being as high as 75% [60]. Furthermore, increased feedstock moisture results in higher H₂ production, and gasification can also be energetically self-sustaining [60,65]. Drying occurs between temperatures of 70–200 °C, with the required heat coming from the partial oxidation reactions. During this stage, the moisture content is reduced to approximately 15% [15]. Pyrolysis occurs between 350–500 °C, with gases/vapors and oils/tars forming. Oxidation and reduction occur concurrently. The oxidation reactions are highly exothermic, which raises the temperature of the gasses to approximately 1100 °C [15], using this heat to maintain the endothermic stage of drying. During reduction, the products of pyrolysis are reduced to permanent gases.

Properties, including surface, size, shape, moisture content, volatile matter, and carbon content, affect the overall gasification process and products. Similarly, the temperature strongly affects the final composition of the products. Increased temperature enhances the yield of hydrogen and the endothermic equilibrium reactions, which may increase the energy output [64]. Temperatures greater than 800 °C lead to high conversion of carbon and high molecular weight hydrocarbons to form gas [15]. However, too high temperatures may lead to high ash content and clinker formation.

5. Fate of CECs during Thermal Treatment

Removal of CECs such as PFAS (perfluoroalkyl and polyfluoroalkyl substances) is critical for the continued beneficial use of BS. Studies have also suggested that thermal

treatment is effective for removing other contaminants, such as microplastics and pharmaceuticals, and personal care products from BS [53,66–68]. Due to fluorine's electronegativity and the chemical stability of fluorinated compounds, PFAS is likely to be the most difficult CEC to destroy/remove/mineralize. This stability and ecotoxicity have earned PFAS the nickname of “forever chemicals” and meant that it is likely to be subject to strong regulations in the near future [69]. In the discussion that follows, we focus on PFAS destruction as a model CEC and emphasize the link to BS moisture content.

Degradation of tetrafluoromethane (CF_4) (a PFAS compound) has the highest degradation temperature of approximately 1200–1400 °C [70]. Given this, it has been suggested that CF_4 should be used as a surrogate for all CEC contamination [71]. Although some authors believe using CF_4 alone may underpredict PFAS destruction efficiency, and multiple surrogate compounds should be considered [72]. A recent review by Garg et al. (2023) covers the current state of treatment technologies for the removal of PFAS from BS [73].

Studies have also shown that the pyrolysis temperature has a significant effect on the bioavailability of nutrients. Mercl et al. (2020) showed that a pyrolysis temperature of 220 °C increased the availability of Ca, K, Mg, P, and S compared to dried BS [74]. However, at 420 °C, all nutrient concentrations were lower than the initial BS, with a further decline in availability with increasing pyrolysis temperature. This is in agreement with Adhikari et al. (2019), who found that pyrolysis operating conditions significantly affected biochar properties (surface area, yield), including P being more tightly bound after increased pyrolysis temperatures [75]. However, CECs such as PFAS may require temperatures up to 900 °C (or higher) for successful removal/mineralization. Hence there is a trade-off between CEC removal and the potential nutrient value of the BS/biochar.

6. Influence of Moisture Content

Initial moisture content is one of the defining factors influencing the energy costs associated with thermal treatment [35], but it can also help expedite the thermolysis of compounds such as PFAS [76]. For example, Altarawneh (2021) modeled the thermal degradation of 1-butanefulfonic acid as a model compound of PFOS and per-fluorinated sulfonic acids in general [76]. Temperatures ranged from 600 to 2000 K. The kinetic model used in this work incorporated four sets of reactions: unimolecular decomposition channels, hydrofluorination, hydrolysis, and fragmentation. Altarawneh's modeling showed that moisture was important in the hydrolysis reactions and that direct moisture addition could expedite the decomposition of larger chain species into smaller ones.

Moisture content can also have an influence on the pyrolysis products' composition and yield. Luo et al. (2017) performed pyrolysis experiments on BS in a bench-scale fixed-bed reactor [77]. The samples were pyrolyzed with steam. Samples were screw conveyed into the reactor, where they dropped onto a ceramic disk, undergoing heating, evaporation, and pyrolysis at 800 °C. The released moisture then formed a steam-rich atmosphere, and in-situ steam gasification of the wet sludge took place simultaneously. Moisture content was found to have a large influence on the proportions of gasification products. Increasing BS moisture (24% to 43%) increased the proportions of gas (39 to 54%), while the proportions of tar (46 to 33%) and char (14 to 12%) decreased. However, high moisture (43 to 62%) decreased the gas content (54 to 49%), while tar and char content increased slightly (up 2.3% and 2%, respectively). Increased moisture content meant more steam was introduced into the gasifier, which enhanced gas yield and accelerated carbon conversion efficiency.

Liu et al. (2014) investigated the influence that residual moisture had during pyrolysis [78]. Three types of samples were analyzed, dry sludge, wet sludge (50.87%), and soaked sludge (sludge that had been dried to remove bound water and then soaked back to the wet sludge MC; hence, most of the moisture should be “free”). Pyrolysis used 2.5 g of sludge sample at 873, 1073, and 1273 K and lasted for 60 min. Gas yield during wet sludge pyrolysis was higher than that of dry sludge, indicating that residual moisture favored gas generation. Additionally, it was found that tightly bound water/trapped water had

a more significant impact on product yields, with bound water being more favorable to char gasification. The reasoning is that bound water had more chance to contact and react with char than free water, which simply surrounds the sludge particles. Moreover, bound water takes longer to evaporate, which prolongs the contact time of steam, sludge, and char particles. H_2 yields were also found to be higher in the wet and soaked sludge compared to the dry sludge, with a minimum difference of 1.2 times. Hydrogen distribution was strongly affected by the state of water, with bound water increasing hydrogen yields more than free water. Bound water results in a char with a higher surface area and additional breakage of C-C bounds, which promotes the conversion of macromolecular organic matter into smaller ones, which can more easily undergo steam gasification.

7. Integration of Drying and Thermal Treatment

A substantial number of papers are available which investigate pyrolysis and gasification of BS, focused on product distribution and compositions, as well as resulting biochar properties. Recent reviews in the area demonstrate the significant efforts being made by researchers [15,28,58,60]. However, research on the integration and combining of drying with pyrolysis and/or gasification is very sparse, despite the importance of MC on process efficiency. The integration of search terms such as thermal treatment, drying, sewage sludge, gasification, and pyrolysis generates only 30–40 results, of which only a handful focus specifically on the integration of these processes. However, additional studies that focus on blending BS with other feedstocks in waste-to-energy (WtE) plants via gasification and pyrolysis were also reviewed, where relevant.

Integrating drying and thermal treatment processes into pre-existing WWTPs is unlikely to be a clonal/cookie-cutter process. Significant consideration of each WWTPs BS properties, including CEC concentration and regulations, as well as local circumstances (plant size, economy, geography, climate, etc.), will ultimately determine which specific drying and thermal treatment technologies are most suitable. Although, as discussed by Spinosa et al. (2011), generalities may be made [79]. Spinosa et al. (2011) suggest that an integrated system should consist of anaerobic digestion, dewatering/drying, and thermal treatment via pyrolysis or gasification. Anaerobic digestion would allow energy recovery via biogas production, which could then be used to help power the dewatering/drying stages. Similarly, it is recommended that any energy produced during thermal treatment (whether that be waste heat reclamation or combustion of product oils/gases) is used to power the dewatering/drying stage.

Figure 3 shows a generic integrated system with various options for heat integration and recovery. One of the key differences in system setup is to either integrate drying and pyrolysis into the same unit, which can improve efficiency and reduce the footprint of the plant, or utilize energy-rich gases from either anaerobic digestion (AD), pyrolysis, or biochar gasification to produce hot flue gas or generate steam to run a separated dryer system. The choice between such systems depends on BS composition, especially the ratio of volatile solids (VS) to total solids (TS) and the conditions in the pyrolysis/gasification that dictate the product (gas, solid, liquid) distributions and yield.

In reviewing this area, three main types of papers were identified. The first involved simulation-based research on combined processes, the 2nd was research using lab-based trials that coupled drying and thermal treatment, and the last was pilot-scale examples of integrated processes. No full-scale examples demonstrating the integration of drying and thermal treatment of BS were identified in the literature.

Lumley et al. (2014) and Patel et al. (2018) developed ASPEN PLUS models simulating various thermal treatments of BS. Lumley et al. (2014) simulated a gasification-based system [80], whereas Patel et al. (2018) examined a pyrolysis system [62]. Lumley et al. (2014) simulated the system using a separate drying and thermal treatment process, whereas Patel et al. (2018) investigated an integrated one. Lumley et al. (2014) design incorporated four processes: convective drying, gasification, gas cooling, and cleaning, and an engine generator [80]. Simulation results showed that 75% of the energy content in the sludge was

required to first dry the dewatered sludge from 80 to 10% and then to heat it to 850 °C. The system recovered heat by recycling all exhaust gas from the engine generator and hot gas from the syngas cooling heat exchanger into the convective dryer, where it made direct contact with the BS. Additionally, a portion of the syngas can be combusted in a utility burner for further dryer heating. The final analysis showed that the system could produce net electrical power, with the dryer's energy needs being met by the recycled exhaust stream and the combustion of 2% of the produced syngas.

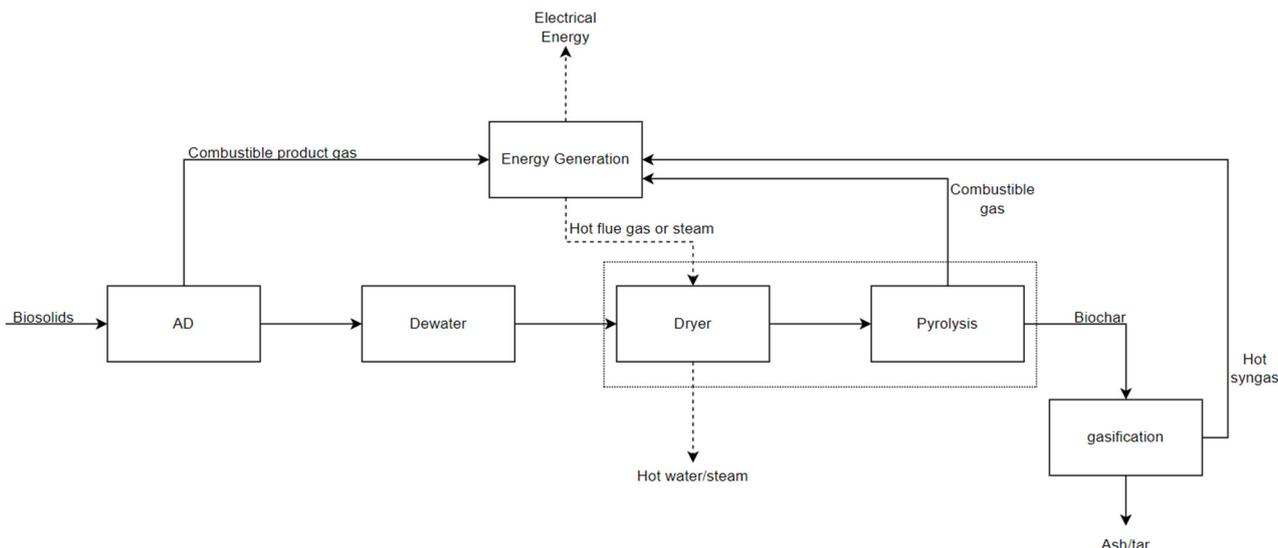


Figure 3. Generic BFD of a combined dryer and thermal treatment system. The dryer and pyrolysis units may be separated or combined into a single system. Various energy-rich gas streams are identified and utilized in an energy generation system that feeds the dryer.

Patel et al. (2018) model had a feed moisture content that ranged from 80 to 20%. The energy was supplied to the dryer/thermal unit via combustion of the py-oil and py-gas generated from pyrolysis. Pyrolysis temperatures from 450 to 850 °C were simulated. The model estimated that BS with less than 50% moisture content did not require external energy for the pyrolysis process. Energy generation (from the production of pygas and pyoil) increased with pyrolysis temperature until it peaked at 650 °C. At higher temperatures, energy generation decreased due to the change in product (py-gas, py-oil) distribution and composition.

A number of researchers have performed pyrolysis/gasification at a lab scale. For example, Gil-Lalaguna et al. (2014) conducted an energetic assessment of two potential treatments for BS: (i) a two-stage process: BS thermal drying + air-steam gasification of the BS and (ii) a three-stage process: sewage sludge thermal drying + pyrolysis of sewage sludge + air-steam gasification of the char derived from the pyrolysis stage [81]. Pyrolysis and gasification were performed experimentally with pre-dried sludge (6.48% MC) as the feed. Pyrolysis experiments were conducted at 530 °C while sludge and char gasification were conducted between 770 and 850 °C. Heating values of the condensed liquid phase were determined via an analytical calorimeter, but it should be noted that the energy potential of the bio-oil resulting from pyrolysis was not considered due to its poor stability and further treatments needed prior to being a viable fuel source. Feed sludge MC was assumed to be 65% for the energetic assessment, which is generally not achievable via traditional dewatering technologies alone, suggesting that additional energy would be required to dry from 80% moisture. The energy balance found that for the first treatment method, the energy contained in the produced gases and vapors if recovered, could be used to cover the energy cost of the thermal drying and gasification stage. However, for the three-stage process, additional energy input was required.

It is important to note that dryer efficiency (i.e., conversion of heat energy to evaporate water) is often assumed to be 100%. Such an assumption is optimistic as it does not consider

practical dryer efficiencies or mass transfer resistance associated with skin formation and shrinking of BS. Such considerations are important.

Tic et al. (2018) proposed a treatment system consisting of a dryer, torrefaction reactor, and gasifier engine [82]. Their study performed drying tests using a commercial scale paddle dryer (2.5 m length \times 1 m width \times 1 m high). Three consecutive drying tests were performed with an initial BS batch size of approximately 1 ton. Table 3 shows the initial and final moisture content of each test and the average consumption of heat energy required to dry the biosolids. As presented in Table 3, there was a clear increase in heat consumption per unit of moisture removed with decreasing final moisture content. This illustrates the influence of increased mass transfer resistance from skin layer formation and shrinkage.

Table 3. Moisture and energy values from Tic et al. (2018) drying experiments.

Parameter	Test 1	Test 2	Test 3	Unit
Moisture content of the feedstock	83.9	53.47	32.13	% (w.b.)
Moisture content of dried Biosolids	53.47	32.13	4.02	% (w.b.)
Average consumption of heat energy	4141	6300	7203	kJ/kgH ₂ O

Biosolid composition and energy content are also important, as shown by Wang et al. (2012), who investigated the combined pyrolysis and gasification of BS at a laboratory scale. Three samples from the same WWTP were collected at different times of the year, and their LHV and MC were determined [83]. Samples were pre-dried to 3 wt% and pyrolyzed between 400 and 550 °C. The py-oil and py-gas were cooled and collected while the biochar was gasified between 800 and 850 °C, with the fuel gas being cooled and analyzed. It was found that the LHV of each sample (14.92, 15.65, 18.45 $\frac{\text{MJ}}{\text{Kg}}$ d.b.) varied due to differences in both sample composition and initial moisture content. Energy requirements were dependent on both the biosolid's LHV and MC. For high LHV (>18 MJ/Kg), the pyrolysis products (i.e., gas) could self-sustain the process, and the biochar could be produced as a useful product without the need for gasification. However, for lower LHV (<15 MJ/Kg) or high MC BS (>80%), then part of the biochar had to be gasified to serve as supplemental energy. Alternatively, if dewatering technologies improve or solar drying is used to reduce the initial moisture content to 65%, the critical LHV was reduced to 10 MJ/Kg.

A similar conclusion was reached by Li and Feng (2018) after they conducted a life cycle assessment of an integrated sludge anaerobic digestion and pyrolysis process [84]. Their study analyzed three different treatment processes (anaerobic digestion-only process, pyrolysis-only process, integrated anaerobic digestion + pyrolysis) and considered sludges with volatile solids/total solids (VS/TS) ratios from 40 to 70%. The initial moisture content for the analysis was taken as 60%, which is significantly lower than the usual 80%. The analysis found that none of the systems could achieve a net energy output until the VS/TS ratio exceeds 50%. For the AD+PY system, net energy was only achieved once the VS/TS ratio exceeded 61%. However, if the initial moisture content was 80%, no surplus energy would be available even if the organic content of the sludge reached 70%.

This dependence on suitable BS composition and MC for energy-neutral processing has also been shown at the pilot scale by Ledakowicz et al. (2019). Their study investigated the integration of a steam-heated twin screw contact (conductive) dryer with a Spirajoule pyrolysis reactor at both the pilot scale (40 kg/h feed) and mobile scale (100 kg/h feed) [34]. In both instances, the drying and pyrolysis processes were separated. Two types of BS were used, one which had been lime treated and one that had not. For both installations, post drying consistency of BS changed from a paste (~80 wt% water) into a mixture of granules (<5–10 mm) and powder. For the pilot scale, dewatered BS (80% MC) was fed to the dryer (40 kg/hr), where its MC was reduced to 10% and 3.5% for sludge with and without lime treatment, respectively. Low-pressure steam (180 C 0.5–0.7 MPa) produced by a gas boiler was used to heat the dryer. Pyrolysis temperatures of 400 °C and 800 °C were investigated, with residence times being 15 or 20 min. Power demands of the pilot plant were between 12 and 15 kW, of which 6 kW was consumed by gear motors/drives and heat losses. The

drying process was the most energy-intensive stage, with energy consumption reaching below 10%, with MC being estimated to be $732 \frac{\text{kW h}}{\text{ton H}_2\text{O evaporated}}$ (or $512 \frac{\text{kW h}}{\text{ton wet sludge}}$ or $750 \frac{\text{kg steam}}{\text{ton wet sludge}}$).

The mobile plant (100 kg/h feed) had a similar process to the pilot plant, with the main process difference being that the hot syngas obtained from the pyrolysis was used for fueling the boiler, which generated the steam for the dryer. Again, no residence time for the dryer is given; however, pyrolysis residence times were increased to 20 and 30 min. Pyrolysis temperature was performed at 850 °C. An increase in residence time from 20 to 30 min increased the gas yield from 31 to 46 wt% at the expense of the oil and char yield, which dropped from 31 to 18% and 38 to 36 wt%, respectively. The full-scale pyrolytic gas had a lower calorific value than the pilot-scale py-gas due to higher mineral composition in the feed sludge. As such, the hot syngas only provided 47 kWh, and an energy deficit of 10–12 kWh was estimated.

A possible solution for BS with low LHV is to blend them with other feedstocks. Several studies have investigated this in waste-to-energy (WtE) plants. Bianchini et al. (2015) proposed using flue gas from a WtE plant to dry BS directly, without a heat exchanger, by introducing milled BS pellets into the flue gas pipe [85]. Dried BS could then be pyrolyzed/gasified. Considering an initial moisture content of 75%, an LHV of 13 MJ/Kg, and a 90% drying efficiency, they proposed that 1 MW of recovered thermal power could be obtained per year. Chiang et al. (2016) studied the co-gasification of biosolids and paper-mill sludge in a commercial-scale plant [86]. They mixed various ratios (1:0, 1:1, 1:2, and 0:1) of sludges before drying and gasification. Initial moisture content varied, depending on the ratio. Drying equipment was heated by 450 °C steam, produced by a boiler fueled by the combustion of the syngas. The gasifier operated at 900 °C. Energetic analysis was performed on the 1:1 ratio, which had an initial MC of 67%, and found that it produced enough energy required for the dryer. The study also found that the calorific value of the gas was slightly lower than that of raw sewage sludge or raw paper, possibly due to difficulty in controlling the gasifier when using mixed sludge optimally. These examples demonstrate that blending an energy-rich feedstock with BS can assist in system energy neutrality.

Logan City Council (2021) in Queensland, Australia, conducted a BS gasification demonstration plant at the Loganholme Wastewater Treatment Plant between January and August 2020 [87]. The plant consisted of a hearth (pyrolizer), thermal oxidizer, heat recovery system, scrubber, and an air/steam stack. Dried BS (10–15% MC, particle size <1–10 mm) was fed at a rate of 480 kg/h (85% of design rate), with the hearth typically running between 500 and 800 °C. While no dryer was part of the demonstration plant, a steam boiler and condensing unit were included to capture and directly measure the heat generation and recovery. Scaling to 100% of the design rate (565 kg/h), the heat recovery was forecasted to be greater than 1.1 MW, which is only marginally larger than the design requirement of 1 MW for the intended belt dryer. However, similar to previous examples, dewatering performance, BS composition, and BS energy content are all critical in driving improved energy efficiency. The plant also saw a 94% reduction of measured PFAS from the BS to the final biochar and a significant reduction in microplastics [87].

The thermal treatment of BS via pyrolysis/gasification is shaping to be a promising solution for removing CECs from BS, thus allowing the resultant biochar to continue to be beneficially reused. However, there is still uncertainty about how economical this solution is due to the high energy costs associated with BS drying and the variance in BS MC and energy content. Many of the studies (both theoretical/simulations and experimental) that have investigated the integration of drying and thermal treatment (pyrolysis or gasification) report that after energy balances/calculations, the energy from the resultant gaseous and liquid products can cover the energy cost required for drying. However, many of these energy balances/calculations are simple and do not consider heat losses, drying efficiencies, or drying resistances of the BS. Furthermore, these studies do not experimentally prove that it is possible to cover the energy needs of drying. Studies that do experimentally determine

this show that generating an energy surplus, or breaking even, is very challenging, and careful optimization will be required. Either additional steps (such as AD and biochar gasification) may be required to obtain energy neutrality, or BS may need to be blended with more energy-rich feedstocks.

8. Conclusions

The reuse of biosolids in agricultural applications is an important part of reducing reliance on synthetic fertilizers, minimizing carbon emissions, and closing the nutrient cycle. Reusing biosolids is being threatened by contaminants such as PFAS and microplastics, collectively known as CECs. As regulations on CECs in biosolids increase, thermal treatment such as pyrolysis and gasification become essential. However, the high moisture content of biosolids (>80%) demands that extensive drying be undertaken to facilitate thermal treatment, a process that consumes significant energy and requires careful optimization when integrated into thermal treatment processes. Both moisture content and temperature influence the effectiveness of CEC destruction, and models that provide internal distributions for both variables would help optimize thermal processing treatments. This review also shows the significant impact of biosolids moisture on the energy products and the product distribution from the thermal treatment process. Moisture has the potential to increase CEC degradation rates. Moisture also results in in-situ gasification and, depending on its magnitude, can lead to higher rates of gas formation. Thus, careful balancing of thermal destruction and energetic product distribution associated with the thermal treatment of biosolids is required. Both integrated and separated drying and thermal treatment processes are identified in this review as viable processing methods. The review shows that the energy and moisture contents of a WWTP's BS are critical factors to consider for the efficient design of integrated processes. However, more research is required to obtain data and suitable models that specifically characterize CEC destruction via pyrolysis and gasification, especially at pilot scales. Careful integration of drying and thermal treatment can overcome energetic challenges. However, the option to blend additional feedstocks to make thermal treatment energetically viable has been shown to be necessary but dependent on BS composition. Further study, including well-validated systems modeling of integrated processes and a more comprehensive analysis of BS compositional variance, is needed to ensure economic viability and drive circular economy objectives.

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Nomenclature

Acronym	Meaning
BS	Biosolids
CECs	Contaminants of Emerging Concern
CE	Circular economies
WWTPs	Wastewater treatment plants
PCPs	Personal care products
PFCs	Perfluorochemicals
MC	Moisture content
PFAS	Perfluoroalkyl and polyfluoroalkyl substances
WtE	Waste-to-energy
AD	Anaerobic digestion
VS	Volatile solids
TS	Total solids
LHV	Lower heating value

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