

# Management of Sewage Sludge from Municipal Wastewater Purification

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Municipal sewage sludge (MSS) is the residual material produced as a waste of municipal wastewater purification. It is a sophisticated multi-component material, hard to handle. For many years, it has been landfilled, incinerated, and widely used in agriculture practice. When improperly discharged, it is very polluting and unhealthy. The rapidly increasing global amount of municipal sewage sludge produced annually depends on urbanization, degree of development, and lifestyle. Some diffused traditional practices were banned or became economically unfeasible or unacceptable by the communities. In contrast, it has been established that MSS contains valuable resources, which can be utilized as energy and fertilizer. Ambitious and costly plans for remediation, the modernization of regulations, collecting and purification systems, and beneficial waste management using a modern approach. The activated sludge process is the leading technology for wastewater purification, and anaerobic digestion is the leading technology for downstream waste. However, biological technologies appear inadequate and hydrothermal carbonization, already applicable at full scale, is the best candidate for playing a significant role in managing municipal sewage sludge produced by big towns and small villages.

Keywords: municipal wastewater ; sewage sludge ; sewers systems

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## 1. Introduction

Few numbers suffice to quantify the relevance of this topic:

- Municipal wastewater (MWW) means the spent water of a community. From the standpoint of source, it may be a combination of the liquid- and water-carried waste from residences, commercial buildings, industrial plants, and institutions, together with any groundwater, surface water, and stormwater that may be present <sup>[1][2]</sup>;
- Sewage sludge is the residue resulting from the treatment of wastewater released from various sources, including homes, industries, medical facilities, street runoff, and businesses <sup>[3][4]</sup>. Municipal sewage sludge (MSS) is the residual material produced as a by-product of MWW treatment plants (WWTPs);
- Municipal sewer systems refers to all sewer mains within the limits of the public right of way, or easements, including maintenance holes and appurtenances <sup>[5][6]</sup>.

In 2020, the volume of MWW generated annually worldwide was estimated to be 360–380 cubic kilometers, predicting a 24% increase by 2030 and a 51% increase by 2050. A little more than half is treated in about sixty thousand WWTPs operating worldwide, while the remaining is released directly into the environment <sup>[7][8]</sup>.

Five years ago, the annual global MSS production rate was estimated to be 45 million tons as dry solid (DS) <sup>[9]</sup>. The result was obtained considering that two billion people were connected to MWW sanitation systems having secondary treatment facilities and quantifying the amount of sludge produced per capita as 70 g of DS per day. In terms of sludge (i.e., the wetted organic and inorganic matter resulting from the treatment of MWW), the amount is from 5 to 10 times higher than in terms of DS <sup>[10]</sup>.

Presently, the total length of the sewer network across the EU has been estimated at around 3.3 million kilometers <sup>[10]</sup>; thus, its total length worldwide is close to one order of magnitude higher (i.e., almost a thousand times the circumference of the Earth). It is also worth mentioning that the EU sewer network length has increased by about 10% during the last four years <sup>[11]</sup>.

The main reason for such a challenging and relevant situation is urbanization, for which there is currently no universal definition. The UN reports figures based on nationally defined urban shares.

## 2. On-Site Treatments

### 2.1. Stabilization and Thickening

Activated sludge treatments <sup>[12]</sup> of municipal sewage followed by thickening and sanitation enable the discharge of more than 99% purified water. A volume reduction of approximately 30–80% can be reached with RMSS thickening. The most

commonly used thickening processes include gravity thickening, dissolved air flotation, and rotary drum thickening. Centrifuge thickening is also becoming more common. The type of thickening selected is usually determined by the size of WWTP, its physical constraints, and the planned further downstream operations. Thickening commonly produces sludge solids concentrations in the 3% to 5% range, whereas the point at which sludge begins to have the properties of a solid is between 15% and 20% solids concentration.

As consequence of the natural fermentation and the presence of pathogenic organisms, RMSS gives rise to offensive odors and health problems. Stabilization is the on-site downstream process that prevent these problems and is usually achieved by chemical, biological, and thermochemical treatments.

The typical chemical stabilization is alkaline stabilization, almost always performed by adding lime, or lime and waste solid materials, to the thickened liquid sludge, to raise the pH to greater than 12 and maintain the resulting materials under these conditions for several days <sup>[13][14][15][16]</sup>. The resulting solid and significantly stable product can be easily and safely transported outside the WWTP for further treatment or final disposal. Recently, ozonation and other advanced oxidation processes (AOPs), such as Peroxone, have attracted attention for the chemical stabilization and reduction in RMSS and as a pretreatment prior to the biological stabilization of RMSS <sup>[17][18][19][20][21][22][23][24][25]</sup>.

Biological stabilization employs microorganisms which develop naturally within the sludge under reactor conditions. It can be conducted under aerobic <sup>[26][27][28][29][30][31]</sup> or anaerobic <sup>[32][33][34][35][36][37][38][39][40][41]</sup> conditions, namely, aerobic stabilization (AS) or AD. In the last case, when the C/N ratio is too high, nitrogen can be added in an inorganic form (ammonia) or in an organic form such as livestock manure, urea, or food waste <sup>[42]</sup>. According to the WEF Manual of Practice <sup>[43]</sup>, AD is generally applied for WWTPs, treating wastewater incoming flows greater than about twenty million liters per day or about 60,000 to 100,000 PE, whereas AS is typically applied in smaller WWTPs. Both AD and AS significantly reduce the amount of raw activated sludge and produce temporarily stabilized wet solid material. However, the energy balance of AS and AD is quite different, because the composition of the resulting solid phase and its dehydration are different. The other relevant difference between AS and AD are investment and management costs, ease of use, and greenhouse gas emissions. Of note is the energy balance which, unlike in AS, a significant amount of biogas composed of 45–60% methane is produced in AD. As a result, AD provides a significant part of the energy required by the wastewater purification process.

Aerobic stabilization resembles the conventional activated sludge process, but excludes wastewater feed and employs longer solids retention times (SRTs). There is no new supply of organic material from the wastewater; therefore, the activated sludge biota begins to die and is used as a substrate by saprotrophic bacteria. This stage of the process is known as endogenous breathing. It has the task of drastically reducing the number of pathogenic organisms and the concentration of organic solids in the sludge, solidifying and temporarily stabilizing the RMSS, which can easily be transported safely outside the IDA for final disposal with or without deriving benefits. An interesting variant of AS is the so-called autothermal aerobic thermophilic aerobic digestion (ATAD) based on the natural phenomenon of the exothermic oxidation of organic carbon with the addition of forced air or oxygen. If the reactor is well insulated, the heat produced is not lost, and the temperature rises to 70–75 °C, with some favorable implications <sup>[44][45]</sup>. Comparisons between AS and AD and between AS and lime stabilization have been reported <sup>[31][46]</sup>.

## 2.2. Dewatering

MSS temporarily stabilized aerobically or anaerobically needs further on-site treatment and dehydration to separate the solid material from an aqueous stream, usually directed to the inlet of the WWTP. The solid resembles a cake, not sliding, and instead forming lumps that can only be transported by a conveyor belt, mechanical earth-moving equipment, or a spade. The first difference between thickening and dewatering is that the former removes free water, increasing the DS content to 4–6%, whereas the latter removes significantly more water content of the sludge to produce a concentrated sludge product with a concentration of SD, generally between 15% and 25%. The second difference is in the way the process is carried out. Dehydration can only be achieved by applying significant mechanical force in a filtration or centrifugation system. Alternatively, the liquid can be allowed to evaporate in ambient conditions (sludge drying lagoons) or either evaporate or drain by gravity through a porous medium on which the sludge is located (sludge drying bed) <sup>[47][48][49]</sup>. Both mechanically dewatered aerobically and anaerobically stabilized MSS contain 75–85% of intracellular, interstitial, and surface water. However, when the treatment capacity of the WWTP is sufficiently high, the AD section can be optimized by pre-treating the thickened raw primary and excess activated sludge by thermal hydrolysis (TH), which obtains better results in terms of biogas production and enhanced dewaterability of the digestate, as discussed by Akajis et al. <sup>[50]</sup>, among others. TH disintegrates organics, allowing for more fermentable organic substances and interstitial water to become free water within the digestate. Typically, the TH reactor operates at 6 bar, 400–440 K, for about half an hour of retention time, followed by treatment in a flash tank operating at atmospheric pressure. The steam produced when flashing was returned for heat recovery by direct contact, preheating the feed to the TH reactor.

## 3. Off-Site Treatments

Stabilized MSS is usually removed from WWTPs for further treatment before beneficial use or for final disposal.

### 3.1. Application to Agricultural Land and Reclamation Sites

In 1991, the U.S. EPA introduced the term “biosolids” for stabilized solid material leaving the municipal WWTPs or POTWs [51]. Australia and New Zealand use the same terminology, which is now familiar and recognizable worldwide. Unlike in the EU, countries that use the term biosolids in their formal documents do not regard this material as waste, but instead as a by-product to be eventually post-treated for recycling. Traditionally, semi-solid and solid materials produced by municipal WWTPs have been widely used around the world as fertilizers and soil improvers, being rich in nutrients, mainly compounds of phosphorus and nitrogen that are bioavailable for vegetation and crop cultivation and carbon-rich substances. However, basic biosolids and stabilized and dehydrated MSS contain various micropollutants, regardless of terminology and regulations. As of 2020, the U.S. EPA [52] classified sewage sludge (biosolids) into Class A—exceptional quality (EQ)—or Class B, based on the specific treatment requirements for pollutants and pathogens, as well as management practices. Minimum requirements for meeting Class A and Class B biosolids are determined by federal regulation 40 CFR Part 503. A 2019 census of the EPA based on the operating POTWS reported that about 51% of produced biosolids found beneficial use as applications to agricultural land and reclamation sites. Similar disposal shares of MSS are maintained worldwide. Despite the increasingly stringent regulations for using biosolids in agriculture and land applications [53], a recent report [54] predicted an increase in this disposal method. However, recent studies [55][56][57][58] have stated that the direct use of sewage sludge and compost containing sewage sludge should no longer be considered a direct source of nutrients and organic matter in agriculture because of their pollutant contents. Agricultural and land applications of biosolids are controversial. The forecast made by the already mentioned market report is likely correct, considering the ongoing evolution of technologies for purifying excessive concentrations of heavy metals [59], microplastics, and other phytotoxic substances [60][61][62]. The practice of land applications for good-quality biosolids remains the most economically feasible and environmentally friendly, and is in harmony with the circular economy and the conservation of non-renewable resources. This practice requires the availability of land not excessively far from the places of production, and is more practicable in countries with a low population density. Composting raw biosolids is the most widely applied technology to enhance the quality of dewatered MSS when used as fertilizer and soil, improving fertility. It is a well-known and globally applied bioprocess allowing the biodegradation of various substrates carried out by a microbial community composed of various populations in aerobic conditions and in a solid state. The exothermic process starts with a mesophilic phase (ambient to 40 °C), proceeds with a thermophilic phase up to about 65–70 °C, and ends with a maturation phase. Completing the first two phases requires about one week, whereas maturation requires up to six months. The main factors that affect the biology of composting are moisture, temperature, pH, nutrient concentration, and oxygen supply. The optimum moisture content is 50–60%; less than 40% moisture may limit the decomposition rate, whereas with an initial mixture higher than 60%, proper structural integrity will not be achieved, and substrates will not decompose well. Temperature rise is important to destroying pathogens, although most mesophiles are killed when it exceeds 70 °C. The optimum pH range for the growth of most bacteria is between 6 and 7.5, and between 5.5 and 8 for fungi. The pH varies throughout the composting steps, and is essentially self-regulating. Carbon and nitrogen are required as energy sources for the growth of microorganisms. The most desirable carbon-to-nitrogen ratio should be 25–35 in the composting mixture by weight. Oxygen concentrations in the composting mass should be maintained between 5% and 15% by volume of gas mass, and should be well distributed. Excessive aeration will compromise the required temperature profile during the process. Composting can be performed in different ways (batch or continuous), using very simple (pile) or sophisticated and automatically controlled reactors (bio cells), depending on the potentiality of the composting plant [63]. As mentioned, aerobically and anaerobically stabilized and mechanically dewatered MSS retain interstitial water. Consequently, the moisture content is higher than that required for composting. For this reason, before composting, woody material is added as sawdust and wood chips that can be recovered and recycled before the maturation phase. In this way, the aeration process is also favored as the porosity of the mass increases. As far as nutrients are concerned, when the C/N ratio is too low, sawdust is added, while the availability of P is always satisfied. The pH value, on the other hand, is within the optimal range for starting the process and, as already mentioned, tends to self-regulate.

### 3.2. Thermal Treatments (TT)

There are a variety of off-site thermal treatments of stabilized and dewatered MSS based on well-consolidated and new technologies which allow for the final disposal with or without beneficial use.

#### TT1: Drying

Drying is a well-known and widely applied unit operation that removes the mixture from a solid substance. For example, a wet solid such as wood can be dried by evaporation of the mixture either in a gas stream or without the benefit of gas to carry away the vapor, although the mechanical removal of such a mixture by expression or centrifugation is not ordinarily considered drying. Analogously, a solid dissolved into an aqueous solution can be dried by spraying it in fine droplets into a hot, dry gas, resulting in the liquid's evaporation. However, evaporation of the solution by boiling it in the absence of gas to carry away the mixture is not ordinarily considered a drying operation. A relevant example is the production of whey protein concentrate (WPC) and whey protein isolate (WPI) by spray-drying the retentate of whey ultrafiltration. Drying can be classified according to the method of operation, i.e., batch or continuous, according to the method of supplying the heat necessary for evaporation of the mixture, i.e., direct driers when the heat is supplied entirely by contact of the gas

with the substance, or indirect driers when the gas and the substance are separated by a wall having good conductivity property; and solar drying, i.e., when the heat is supplied entirely by exposing the substance to solar radiation.

Furthermore, classification can consider the nature of the substance to be dried: rigid solid, flexible material, granular solids and crystals, thick paste, thin slurry, or a solution. The residual mixture content after drying is typically 8–10% [64]. The water industry has practiced the drying of stabilized and mechanically dewatered MSS for many years to further reduce the volume of the product, making its storage, transportation, packaging, and retail easier. Commercial and legal pressures have also focused on the final product and its use, i.e., for producing EQ biosolids and as a pretreatment for further intensive thermal process [65][66][67][68][69].

#### TT2: Torrefaction

Torrefaction is a thermochemical process performed at 200–300 °C, at atmospheric pressure, in an inert or low-oxygen-content flue gas or superheated steam conditions, and at a relatively low residence time. Dry torrefaction transforms biomass into a superior handling, milling, co-firing, and clean renewable energy into a carbonaceous and hydrophobic solid [70]. Torrefaction enhances feedstock's properties in several ways to enable its use as a direct fuel: reduction in moisture, increase in energy density, reduction in the O/C and H/C ratio, increase in heating value, and improved ignitability and reactivity of the processed fuel. Historically, development of the torrefaction process only began with coffee production in the late 19th century, as documented in the first patents by Thiel (1897) and Offrion (1900). Additional historical information is available in the literature [70][71]. The most recent efforts and current research and development on torrefaction are extensive, with more than two thousand papers assessed as part of the research, published in the last ten years. The main product of the torrefaction process is a coal-like solid, while liquids and gases are produced as by-products. Torrefied sewage sludge is recognized as a better solid fuel than the dry sewage sludge, and it is also beneficial for subsequent thermochemical processes and for producing soil improver and adsorbent material [72][73][74][75][76][77].

#### TT3: Hydrothermal Carbonization and Hydrothermal Liquefaction

Hydrothermal carbonization (HTC) produces carbonaceous material, hydrochar (HC), from very wet biomass, such as AS and AD sewage sludge. HTC had fallen into relative obscurity after the initial investigations and first description by Friedrich Bergius in 1913. Since then, research activity was slowly initiated to understand natural coal formations, until recent studies on HC chemistry and applications in innovative materials and soil quality improvements revived interest [78]. As described by the researchers in a recently published paper [79], HTC is performed at 180–250 °C, approximately the vapor pressure of water under the operating temperature. Typical water-to-biomass ratios range from 5 to 10, and the residence time ranges from 0.25 to 2 h. It works in batch or continuous mode. In addition to hydrochar, HTC produces a large volume of process water (PW), plus a small amount of a CO<sub>2</sub>-rich gas phase. Water acts as a solvent and reaction medium, and the mechanisms mainly involve decarboxylation, dehydration, and polymerization (FZ). During the process, the ionic product of water increases, whereas the dielectric constant decreases. Consequently, water acts more as a non-polar solvent [80]. Furthermore, it has been found that, whatever the biomass, the time course of electrical conductivity follows a unique law, unquestionably corresponding to the evolution of solid-phase carbon content [81]. HTC is still in an evolutionary phase, even though in Europe, many pilot plants and some full-size plants have been built and managed, and a few years ago in China, a full-size plant was built with German technology and is operational for the final disposal of MSS with the recovery of commercial materials. HTC technology is emerging for MSS management, with several favorable implications. The primary and excess sludges, well thickened or slightly dehydrated, can be hydrothermally carbonized directly in situ, i.e., inside the WWTP, due to the favorable ratio between water and organic material. The most favorable conditions apply to small- to medium-sized WWTPs, where an AD section is economically impractical, if not difficult to manage. In this case, the integration of WWTP with an HTC section was drastically reduced [79] the quantity of sludge, i.e., the mechanically dewatered and aerobically stabilized sludge, and which must be disposed of definitively outside the WWTP. Without the HTC section, the sludge to be disposed of has a mixture of about 80%. Considering that the HC yield as DS after the HTC treatment is in the range of 55–65% [82] and the residual mixture content of the mechanically dewatered HC is 30–35% [83][84], it amounts to 16–20% of the solid waste that a small–medium WWTP must dispose of paying, an average of EUR 200 per tons. The possibility of obtaining a solid residue with such a low moisture content depends on the granulometric and hydrophobic characteristics of the HC, which appears as a very hard and finely divided brown solid. HC is sterilized and free of organic volatiles compounds, its moisture content can be further reduced by temporary thin-layer exposure to air or by using warm exhaust air, if available, in a free drying unit. To better quantify the advantages of the direct on-site treatment of RMSS by HTC, researchers took 4000 T/a of AS and mechanically dewatered MSS produced by the “Ponte Rosarolo” WWTP in L'Aquila, Italy, as a case study [85]. A saving of between 80% and 84% was obtained, i.e., EUR 640,000–672,000/a, corresponding to about 50% of the overall purification costs of the WWTP. The press-filtered HC obtained by the HTC of primary and excess sludge produced by the activated sludge process exhibited significant favorable properties, being biologically stable (sterilized).

Furthermore, HTC degrades microplastics (MPs) [86], pharmaceutical and personal care products (PPCPs), and other persistent organic pollutants such as polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs) [87], thus giving a significantly better material in comparison with the digestate.

The on-site HTC of RMSS results in a very efficient stabilization and sanitation treatment. After dewatering, the cost for the eventual disposal of the HC with the residual mixture content is much lower than the cost for the disposal of the AS and mechanically dewatered SS. For the potentiality of the already mentioned WWTP "Ponte Rosarolo", a reactor of two cubic meters enabled a residence time of almost two hours, more than required to account for unperfected mixing. In addition, three pumps, a double heat exchanger, a small boiler for steam production, and piping had to be installed in place of the aerobic stabilization tank with its ventilation system. An investment of EUR 1.5–2 million has been estimated, including design, construction supervision, and assembly, corresponding to a pay-back time of about four years. In contrast, AD is not economically feasible. Researchers did not find any report, paper, or research discussing similar findings. Small–medium WWTPs are numerous, but their full treatment capacity is significantly smaller than that of WWTPs serving big cities. Only little information is available on treating AS and mechanically dewatered sewage sludge, which is usually performed in large-scale facilities for final disposal. However, HTC has been considered to stabilize further and dewater digested sludge other than facilitating phosphorous recovery and as an on-site pretreatment or post-treatment of AD to improve the methane potential of the primary and excess sludge [88][89][90][91][92][93]. Researchers also found a laboratory research paper on the direct application of HTC to the natural mixture of human feces and urines [94]. Hydrothermal carbonization is effective in the disinfection of sewage sludge [95]. Therefore, it attracts attention to the treatment of any biologically derived MSS before their beneficial use or final disposal in the environment. Many companies, researchers, stakeholders, and other entities involved in MSS management are considering the design, development, and construction of high-potentiality plants for the hydrothermal carbonization of AS and AD MSS produced by all operating WWTPs in a given area, as an alternative to more conventional treatments [96][97]. In such a circumstance, the PW cannot be recycled to the inlet of a WWTP and needs to be purified. Economic feasibility is only achievable for high potentialities. The necessary authorization for the construction and operation of this system is still difficult to obtain, considering that the regulations in force in the EU and other countries do not yet explicitly include HTC among the applicable technologies. However, many projects concerning research and technological development for applying HTC to treat MSS have received significant public funding. When the operating temperature of the hydrothermal treatment is in the range of 300–360 °C under pressurized water with enough time (0.25–1 h), the treatment is known as hydrothermal liquefaction (HTL), or direct liquefaction, because it enables the conversion of most of the biomass into a liquid bio-oil by breaking down the solid biopolymeric structure into a mainly liquid components. When HTL is applied to MSS [98][99][100][101][102], the yield in bio-oil is 35–50% of the MSS as DS, depending on the severity of the treatment. It is considered a promising green method for the beneficial conversion of sewage sludge. If the operating temperature is higher than the critical temperature of the water, i.e., 374.1 °C, and the operating pressure is higher than the critical pressure of water, i.e., 21.83 MPa, the treatment is usually termed supercritical water gasification (SCWG). It is accepted as a promising technology for sustainable MSS beneficial disposal [103][104][105][106][107] because it eliminates the need for costly water reduction and drying processes before disposal by conventional methods. Under typical pressure (25–30 MPa) and temperature (450–550 °C) operating conditions, the decomposition of organic and metallo-organic compounds takes place in a few minutes, giving rise to a well-mixed gaseous mixture rich in hydrogen and methane. In fact, in such conditions, water behaves as a completely non-polar solvent, capable of solubilizing organic substances but not inorganic substances. From this point of view, SSCWG produces a solid residue similar to that produced by combustion, i.e., a very high phosphorus concentration. Notably, modern activated sludge processes remove 90% of phosphorus in the treated water by accumulating it in the sludge, and the HTC of AS and AD-stabilized MSS transfers 93–97% of the phosphorus to HC in the form of organic and inorganic phosphorus (ARB). In these circumstances, the concentration of phosphorus in the solid inorganic residue produced by the SCWG is comparable with that type of phosphate rock. Consequently, SCWG coupled with HTC could represent an interesting alternative to incineration when phosphorous recovery must be achieved. The catalytic gasification of sewage sludge in supercritical water enhanced phosphorous recovery and hydrogen production [108][109]. The final and complete destruction of the organic pollutants in the gaseous mixture has also been considered as an alternative, using supercritical water oxidation (SCWO) technology. Since 2001, several commercial-scale SCWO plants have been installed to treat MSS with solid-containing rates of 7–8 wt.%. Nevertheless, the three key problems concerning corrosion, plugging triggered by salt precipitation, and high running cost still exist, even making some commercial-scale SCWO plants inactive [110].

#### TT4: Pyrolysis

The production of charred matter always involves a thermochemical conversion process. The decomposition of organic material under the influence of heat in a gaseous or liquid environment, without the involvement of further reactants (oxygen), is called pyrolysis: from the Greek words 'pyr', meaning fire, and 'lysis', meaning dissolution [78]. Typically, the pyrolysis or carbonization of woody biomass is carried out at atmospheric pressure by heating the material to 281 °C to cause the breakdown of heavy organic molecules and trigger a series of exothermic reactions capable of obtaining further heating of the mass up to 500–600 °C. Under these conditions, the main product is anhydrous carbon with a high energy density. At the same time, a gas is produced that contains all the water initially present in the feed, the volatile and less volatile organic substances resulting from the thermal splitting of the larger molecules into coal, and compounds which, at room temperature, have a bituminous appearance [111][112]. When the moisture content of the feedstock is not excessively high, i.e., less than 35%, the combustion of the gaseous phase may be sufficient to support the endothermic phase of the pyrolysis process. At the same time, it supports the elimination of a gaseous effluent that should otherwise be re-condensed to avoid atmospheric pollution. A more general and current definition is that pyrolysis refers to the decomposition of organic matter at elevated temperatures (300–1300 °C) in the absence of oxygen and under

substantially dry conditions. The operating pressure is usually atmospheric, although vacuum pyrolysis (0.1–0.2 bar) and pressurized pyrolysis (5–20 MPa) have been studied and reported. In all cases, oxygen-free conditions are maintained [47]. The pyrolysis of digested and mechanically dewatered MSS has been widely studied and described in the literature [113][114][115][116][117][118][119]. However, such MSS must be further dehydrated by drying before pyrolysis treatment to avoid the need for recondensation with the formation of a highly polluting organic bituminous phase, which is very difficult to handle due to its unfavorable rheological properties. In contrast, pyrolysis can be applied to mechanically dehydrated HCs because the concentrations of bituminous and other organic compounds in the gaseous phase are about double that of water; consequently, it can be burned, obtaining a combustion gas that can be used to sustain the endothermic phase of the process entirely before it discharges safely into the atmosphere [120]. In summary, the on-site HTC of AS and mechanically dewatered MSS could be followed by the off-site pyrolysis of the HC produced by all the WWTPs of a given area in order to minimize further the unit amount of the solid by-product, charcoal, which can be further processed, as discuss in the following sections. It would be interesting to compare the on-site AD of primary sludge and that in excess with the on-site HTC of the same material followed by off-site pyrolysis in a plant serving WWTPs operating in a specific area. The quantity of coal is much lower than that of the mechanically dehydrated digestate and the HC with its residual content of the mixture, although the quality is much higher for the subsequent beneficial waste-to-energy and waste-to-energy treatments.

#### TT5: Gasification

As already mentioned, biomass can be gasified in the absence of oxygen by pyrolysis or in a supercritical water environment. However, typical gasification is a process that converts carbonaceous organic or fossil-based materials at high temperatures (>700 °C), without combustion, with a controlled amount of oxygen or vapor, into carbon monoxide, hydrogen, and carbon dioxide. Carbon monoxide reacts with water to form carbon dioxide and hydrogen via a water–gas displacement reaction ( $\text{CO} + \text{H}_2\text{O} \rightarrow \text{CO}_2 + \text{H}_2$ ). Modern membranes can separate hydrogen from carbon dioxide and small amounts of impurities [121]. In general, biomass does not gasify as rapidly as coal, producing other hydrocarbon compounds in the gas mixture leaving the gasifier; this is especially true when no oxygen is used. Consequently, a further step must be taken to reform these hydrocarbons with a catalyst to produce a clean syngas mixture of hydrogen, carbon monoxide, and carbon dioxide. Thus, just like in the gasification process for hydrogen production, a displacement reaction step converts carbon monoxide into carbon dioxide. The hydrogen produced is then separated and purified. Gasification is a well-known technology and has been considered for the beneficial treatment of MSS. The results have been reported and discussed in detail in several articles [121][122][123][124][125]. It is worth emphasizing that its economically feasible applicability is only obtained for high potentiality. Furthermore, some pretreatments, such as drying, torrefaction, pyrolysis, or hydrothermal treatments of raw material, are usually applied before final gasification.

#### TT6: Incineration

Incineration is a combustion process aiming to destroy or minimize waste, particularly that produced in urban areas. The destruction is a consequence of burning the organic fraction of the waste with an oxygen concentration well above the stoichiometric one to ensure the reaction is complete. It is a typical waste-to-energy (WTE) operation, converting the waste into ash (primarily inorganic compounds), flue gas to be cleaned before discharging in the atmosphere, and heat, often used to generate electric power. The first incineration facility for waste disposal was built in the United Kingdom about 150 years ago [126]. Since then, incinerators have been built and operated in the United States and other developed and developing countries, mainly for WTE from municipal solid waste (MSW) and for destroying hazardous waste. The incinerator's operating temperature must be above 850 °C, and large plants' treatment capacities can reach about three thousand tons per day. However, much smaller and mobile incinerators are in operation in some cases. One particular case of incineration uses a cement kiln where the ash is incorporated into the mass of a Portland cement clinker [127]. The effect of phosphorus and iron on the composition and property of the product has recently been quantified and discussed [128]. The application of incineration for the final disposal of MSS has been widely considered worldwide, with the exclusion of undeveloped countries and some recent adverse opinions [129]. It has been reported that sludge with a moisture content of no more than 40% can stably burn in a fluidized bed without any auxiliary fuel input [130]. Sewage sludge incineration coupled with phosphorous recovery is the most preferred MSS treatment in Germany, Japan, and many other industrialized and wealthy countries lacking phosphorous mineral deposits. A recent report by the German Environment Agency [131] accurately analyzed technical, economic, environmental, and health issues related to sewage sludge disposal. Notably, the report considered the unavoidable problems created by land applications of only biologically treated sewage sludge containing ingredients of concern such as pharmaceutical residues, hormonally active substances, nanoscale substances, microplastics, and various pathogens. These substances accumulate in sludge and can only be destroyed by proper thermal treatments. About ten years earlier, Bauerfeld et al. [132] stated that with the planned new restrictions, sewage sludges might not be suitable for agricultural reuse any longer. As a 2020 update, Schnell et al. [133] highlighted Germany's mono-incineration situation, the disadvantages of co-incineration, and possible alternative thermal technologies. The choices made by Germany on the use of the MSS are only partially applicable to other EU member states, and are different from those in other countries with a high rate of socio-economic development. In the case of developing countries, these are not applicable due to a lack of resources and limited technical capacity to manage complex plants. The fact remains that biological treatments alone and subsequent insertion or spreading in or on the soil is not sufficient to avoid irreversible environmental damage and severe sanitation problems.

Process integration which involves the interconnection of two or more unit operations to treat complex raw materials, e.g., MWW and related MSS, is an attractive strategy because, theoretically, it could recover energy and separate all the components. The technologies currently available and those under development <sup>[134]</sup> could enable reaching the objective, but the limit lies in the economic feasibility deriving from the need to recover the necessary investments and bear the operating costs with the commercial value of the products. A biorefinery implies process integration, but used when the aim is the recovery of chemicals to be further used as renewable raw materials for synthesizing commercial products. There are numerous contemporary examples of consolidated on-site process integration and biorefinery activities applied at full scale within large WWTPs. In addition, numerous laboratory studies and research and development activities at pilot plants are ongoing worldwide, along with some advanced projects for converting WWTPs into water resource recovery facilities (WRRFs). The hybrid ASP–AD process with electrical energy and heat production is the most relevant example of process integration. Nitrogen (N), phosphorus (P), carbon (C), and related compounds are the most interesting for quantity and usability <sup>[10]</sup>. The recovery and upgrading of dewatered digestate or AS (biosolids) usable for agricultural and land application are significant examples of first-generation biorefinery activities. However, this practice is questionable with the evolution of legislation. Some EU member states have already banned or drastically limited the use of biosolids for agricultural purposes, and others are expected to follow this route. However, as already mentioned, the EU is heavily focused on recovering P from MSS because of its uneven distribution across Earth, and its use in various applications. Notably, P concentrations in MWW range from 5 to 20 mg/L, whereas in purified water, P concentrations must be much lower, usually less than 1 mg/L. The difference is accumulated almost entirely into the MSS-derived HC or the digestate. It may be calculated that the present potential for the annual recovery of phosphorous on a global basis amounts to 5.6 million tons. This annual amount could reasonably rise to more than 9 million tons in thirty years, more than one-third of the current production from non-renewable mineral reserves. On-site pre- and post-treatment of sludge produced by ASPs by chemical (ozonization or similar) or thermochemical (thermolysis, torrefaction, pyrolysis, and HTC) processes for enhancing their methane potential are examples of contemporary full-scale applications of process integration or biorefinery. The ongoing conversion of Avedøre WWTP to a WRRF by 2025 (a VARGA project in Copenhagen, Denmark) <sup>[135]</sup> is a significant example of the ongoing development of a full-scale biorefinery. The research and development activities discussed by Dufour et al. <sup>[136]</sup> and other researchers <sup>[137][138][139]</sup> are examples of future predictable second-generation biorefinery applications.

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