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# Environmental and economic assessment of waste-to-energy treatment paths for organic waste

# Dissertation submitted by Felix Mayer for a Doctoral Degree (Dr. rer. nat.)

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#### **Declaration**

I declare that the dissertation here submitted is entirely my own work, written without any illegitimate help by any third party and solely with materials as indicated in the dissertation. I have indicated in the text where I have used texts from already published sources, either word for word or in substance, and where I have made statements based on oral information given to me. At all times during the investigations carried out by me and described in the dissertation, I have followed the principles of good scientific practice as defined in the "Statutes of the Justus-Liebig University Gießen for the Safeguarding of Good Scientific Practice".

Darmstadt, 05.08.2021

Felix Mayer

#### **Executive Summary**

Waste wood, food waste, the organic fraction of municipal solid waste (OFMSW) and sewage sludge represent the main organic waste streams, which are disposed of as waste in Germany. Given that reuse of these four waste streams is not applicable, the *European Waste Framework Directive* suggests recycling, recovery or ideally a combination of both. The respective treatment paths would return nutrients to the nutrient cycle (recycling) and/or exploit energy from the waste substrate (recovery). Within this work, organic waste was primarily considered as a resource for thermal and electrical energy production and the treatment paths were outlined as waste-to-energy systems. On a meta level, this work aimed at finding preferable treatment paths for the respective substrates. According to the mindset of a circular economy, a preferential treatment path is characterized by minimum impairments to the human health and the ecosystem quality, while considering the entire life cycle of the waste treatment. As a subordinate criterion, it must be ensured that the economic feasibility is not compromised. To quantify these aspects, the methodologies of environmental life cycle assessment (eLCA) and levelized costs of exergy (LCOE) were utilized.

When outlining potential treatment paths for the four organic waste streams, a treatment according to five principal technologies was considered. Herein two conventional technologies, i.e., anaerobic digestion and incineration and three "alternative" technologies, i.e., hydrothermal carbonization (HTC), pyrolysis and gasification were comprised. The technologies were either considered individually or in a cascade. In total 35 potential treatment paths were designed, inventoried and quantified with regards to their environmental and economic burdens.

The results showed that alternative treatment technologies did not hold the potential to reduce environmental or economic impacts considerably and conventional technologies stated suitable solutions to comply with the demand in a high-quality treatment of organic waste. For waste wood, incineration was an apt solution, for food waste anaerobic digestion or a combination of anaerobic digestion and incineration was preferable, for *OFMSW*, either anaerobic digestion or incineration were applicable and for sewage sludge, a combination of anaerobic digestion and incineration stated an acceptable treatment path. The economic feasibility was given for all solutions. Alternative treatment systems were only found to result in increased benefits for niche solutions.

#### Kurzfassung

Altholz, Speisereste, Bioabfall und Klärschlamm stellen Hauptfraktionen des organischen Abfalls in Deutschland dar. Entsprechend der europäische Abfallrahmenrichtlinie werden hierzu Recycling, Verwertung oder idealerweise eine Kombination aus beidem empfohlen. Die Behandlungspfade führen dabei Nährstoffe in den Nährstoffkreislauf zurück (Recycling) und/oder setzen die Substrate in Energie um (Verwertung). In dieser Arbeit wurden organische Reststoffe primär als Ressource für die thermische und elektrische Energieerzeugung betrachtet und Behandlungspfade als Waste-to-Energy-Systeme gestaltet. Das Hauptziel dieser Arbeit bestand darin, vorteilhafte Behandlungswege für diese Substrate zu finden. Im Sinne einer Kreislaufwirtschaft zeichnet sich ein vorteilhafter Behandlungspfad durch minimale Beeinträchtigungen der menschlichen Gesundheit und der Umwelt aus, wobei der gesamte Lebenszyklus der Abfallbehandlung zugrunde gelegt wird. Als untergeordnetes Kriterium muss sichergestellt werden, dass die Wirtschaftlichkeit nicht beeinträchtigt wird. Um diese Aspekte quantifizieren zu können, wurden die Methoden der Ökobilanzierung (eLCA) und der Gestehungskosten von Exergie (LCOE) angewandt.

Bei der Skizzierung möglicher Behandlungspfade wurden insgesamt fünf Technologien betrachtet. Diese umfassten zwei konventionelle Technologien, i.e. anaerobe Vergärung und Verbrennung, und drei "alternative" Technologien, i.e. hydrothermale Karbonisierung (HTC), Pyrolyse und Vergasung. Die Technologien wurden entweder einzeln oder in einer Kaskade angewandt. Insgesamt wurden 35 mögliche Behandlungspfade entworfen, inventarisiert und hinsichtlich ihrer ökologischen und ökonomischen Auswirkungen quantifiziert.

Die Ergebnisse zeigten, dass alternative Behandlungstechnologien die ökologischen oder ökonomischen Auswirkungen nicht erheblich reduzieren konnten, und konventionelle Technologien geeignete Lösungen darstellten, um den Anforderungen an eine qualitativ hochwertige Behandlung von organischen Abfällen gerecht zu werden. Für *Altholz* wurde die Verbrennung als passende Lösung angesehen, für *Speisereste* war die anaerobe Vergärung oder eine Kombination aus anaerober Vergärung und Verbrennung vorteilhaft, für *Bioabfall* waren entweder anaerobe Vergärung oder Verbrennung günstig und für *Klärschlamm* stellte eine Kombination aus anaerober Vergärung und Verbrennung einen gangbaren Behandlungsweg dar. Bei allen Lösungen war die wirtschaftliche Machbarkeit gewährleistet. Alternative Behandlungssysteme führten nur innerhalb von Nischenlösungen zu einem erhöhten Nutzen.

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- Stobernack, Niklas; Mayer, Felix; Malek, Christian; Bhandari, Ramchandra (2020): Evaluation of the energetic and environmental potential of the hydrothermal carbonization of biowaste: Modeling of the entire process chain. In: *Bioresource technology* 318, S. 124038. DOI: 10.1016/j.biortech.2020.124038.
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#### **List of Abbreviations**

AbfKlärV Sewage Sludge Ordinance (dt.: Klärschlammverordnung)

AbwV Wastewater Ordinance (dt.:Abwasserverordung)

BImSchV Federal Emission Control Act (dt.: Bundesimmissionsschutzverordnung)

BioAbfV Biowaste Ordinance (dt.: Bioabfallverordnung)

CHP Combined Heat and Power
COD Chemical Oxygen Demand

DepV Landfill Ordinance (dt.: Deponieverordnung)

DM Dry Matter

DüMV Fertilizer Ordinance (dt.: Düngemittelverordnung)
DüV Fertilization Ordinance (dt.: Düngeverordnung)

EEG Renewable Energy Sources Act (dt.: Erneuerbare-Energien-Gesetz)

EKC Environmental Kuznets Curve

eLCA Environmental Life Cycle Assessment

FM Fresh Matter

GWP Global Warming Potential
HTC Hydrothermal Carbonization

LCI Life Cycle Inventory

LCIA Life Cycle Impact Assessment

LHV Lower Heating Value

LCOE Levelized Costs of Exergy

MFE Mineral Fertilizer Equivalent

NMVOC Non-Methane Volatile Organic Compounds
OFMSW Organic Fraction of Muncipal Solid Waste

oDM Organic Dry Matter

P Phosphorous

PE Person Equivalents

SCR Selective Catalytic Reduction

SNCR Selective Non-Catalytic Reduction

ThOD Theoretical Oxygen Demand VOC Volatile Organic Compounds

WtE Waste-to-Energy

WACC Weighted Average Cost of Capital

WWTP Wastewater treatment plant

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	Chapter 1
Chapter 1	

**General Introduction** 

#### 1.1 Sustainability and waste management

Improper or malfunctioning waste management may compromise social well-being and negatively affect the earth's spheres by emissions and natural resource exploitation. To curtail negative social and environmental implications, the ideal treatment of waste streams results in a minimum interference with the biosphere and a maximum benefit for the society. At the same time, the waste management sector is subject to economic boundary conditions and treatment options are restricted by the willingness-to-pay of the involved parties. This maneuvers the waste management sector in the tension field of environmental, social and economic sustainability aspects. An improvement on the level of all three sustainability aspects is desirable but generally mutually exclusive. Still, an advancement of the waste management sector towards a sustainable development can only be considered as such if the sum of the sustainability aspects is enhanced. This requires an understanding of a sustainable development and how to measure it.

In the scientific scope, the concept of a sustainable development is often described as "meeting the needs of the present without compromising the ability of future generations to meet their own needs" (UNITED NATIONS BRUNDTLAND COMMISSION 1987). As simple as the concept appears, as complex are its implications to measure it. Within the waste management sector each of the three sustainability pillars is subject to a multitude of subjacent aspects (Appendix A). To understand whether a change in the waste management sector may be considered as a step towards increased sustainability, all facets would have to be holistically assessed, evaluated and put into perspective. Lastly, this is an impossible task, mainly because (i) not all mechanisms of anthropogenic actions are understood, (ii) not all impacts can be described quantitatively and (iii) putting sustainability aspects in relation to one another always introduces a strong subjectivity by the practitioner. Therefore, it must initially be determined, which aspects are of the highest relevancy for the waste management sector.

#### 1.2 Waste Hierarchy and circular economy

As hypothesized in the Environmental Kuznets Curve (EKC), environmental well-being is at first largely subordinated to economic growth, up to a point, at which certain economic needs within a population are satisfied and environmental management is allowed for (STERN ET AL. 1996). Alongside other developed countries, Germany has long since crossed this turning point and is thus capable of taking effective measures to mitigate anthropogenic environmental impacts. Although the concept of the EKC was yet only proven for emissions that affected the residentiary population directly, the growing awareness to understand the environment and respective ecosystems as a global complex, may lead to a paradigm shift. A prime example

for externalized environmental pollution on a global scale is the emission of greenhouse gases, which are agreed on, to alter global climate conditions. By formulating objectives to reduce greenhouse gases on a global level (Kyoto-Protocol and Paris Agreement), a European level (EU climate target) and on a national level (e.g. German climate action plan) it becomes apparent that also externalized damages are increasingly targeted.

The developments outlined in the EKC also took root in the waste management sector in the European scope. The central statute in the field of waste legislation, the European Waste Framework Directive, states its purpose as "protect[ing] the environment and human health" (Article 1, Directive 2008/98/EC). Thereby it shifts the focal point of waste management towards environmental and social sustainability. While the directive also declares that economic feasibility must be maintained it clearly implies that waste management should not be conducted according to the most economic option only. These principles were equally harmonized in the German legislation in the realm of the Circular Economy Law (KrWG 2012).



Figure 1: Waste hierarchy according to the Waste Framework Directive (Directive 2008/98/EC)

To reach the designated purpose of the *European Waste Framework Directive*, the concept of a circular economy is brought forth. In contrast to a linear economy, it demands to decelerate and close material and energy loops and to minimize the consumption of natural resources. Even though the term circular economy suggests that waste treatment can be designed as a completely reversible circular process, it should be understood as an idealized idea or even romanticized concept. Any circular waste management concept will always contain traits of a linear economy. Applying the second law of thermodynamics to waste management, shows that - in order to maintain a fully reversible cycle - the increase of the entropy in the waste production must either be compensated by large amounts of external energy or by deviating from a "perfect" cycle (KORHONEN ET AL. 2018).

Therefore, the aim must be a maximum convergence with this circle. Recommended measures to reach this aim were designed in the form of a waste hierarchy. The *European Waste Frame Work Directive* identified five steps in this context, whereby step 1 represents the most desirable and step 5 the least desirable option. Namely, they are 1.) prevention, 2.) reuse, 3.) recycling, 4.) recovery and 5.) disposal. The hierarchy is illustrated in Figure 1 and described in

detail in Appendix B. Within the waste hierarchy reuse appears to be prioritized over recycling and recovery. A strict order is however not applicable and according to Article 4 (2) of the Directive 2008/98/EC, this initial prioritization is not meaningful for all waste streams equally. A deviation from this hierarchy treatment becomes admissible, if the "overall environmental outcome" is enhanced along steps 2-4 of the waste hierarchy. This has to be justified by life cycle thinking.

#### 1.3 Life cycle thinking in the realm of waste management

To determine whether the overall environmental impacts can be reduced, a quantitative approach based on a scientifically sound foundation is necessary to determine the most environmentally friendly solution. To comprehensively assess treatment options, a life cycle approach becomes necessary. This implies that all environmental impacts must be accounted for along the life cycle of a waste material. The methodology to conduct such an assessment is referred to as environmental life cycle assessment (eLCA) and was formalized by international standards in 1997. Today the ISO 14040:2006 and ISO 14044:2006 represent the norms for undertaking high quality and internationally acknowledged eLCA.

Four steps are contained in the eLCA: 1.) definition of the goal and scope, 2.) life cycle inventory, 3.) life cycle impact assessment and 4.) interpretation. Figure 2 shows the framework of an eLCA according to the respective ISO norm. In Appendix B the steps of an eLCA are outlined. The workflow of an eLCA is an iterative process and steps 1-4 interact with each other. Each step must constantly be refined to achieve compliance with the goal and scope, which was initially set for the study.

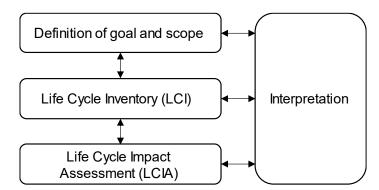


Figure 2: Steps of the eLCA according to the ISO 14040:2006

#### 1.4 Scope and objectives of this work

Over the last decades, Germany revolutionized its waste management sector in accordance with the European waste framework directive (Appendix D). Therein the principles of the di-

rective were assimilated and sound approaches for the collection and treatment of the generated waste were identified and implemented. In the light of technological advances, new scientific findings and changing framework conditions, however, waste management practices need to be constantly reevaluated and adjusted to comply with the aims of the waste framework directive. This work focused on the reevaluation of the treatment paths for organic waste streams under the consideration of conventional and "alternative" treatment paths.

Currently, municipal waste is generated at a rate of ~600 kg per person and year (DESTATIS 2020a). The second largest fraction after residual waste is herein source segregated organic waste, which accounts for >20% of the municipal waste. The actual share of organic waste is deemed to be much higher, as it may be assumed that not all organic waste is accurately source segregated (e.g., KNAPPE ET AL. (2019a)). The high annual throughput indicates a high relevancy for a treatment path with a strong performance. A major problem of organic waste lies in the heterogeneity of the material with a multitude of sub-fractions. These fractions differ in their chemical composition and heating value. They consist of combustible and inert waste, biodegradable and biologically stable waste fractions and varying levels of potentially toxic elements. The nature of the diverse waste streams suggests that tailor made treatment paths become necessary for the individual waste materials. A "one size fits all" approach may not be expedient or even applicable. In compliance with the Waste Framework directive, the methodological approach of life cycle thinking becomes necessary to determine beneficial treatment paths for organic waste streams.

The investigated organic waste streams in this work were the dominant fractions found in organic waste and were (i) the organic fraction of municipal solid waste (OFMSW), (ii) food waste (iii) waste wood (classes AIII and AIV) and (iv) sewage sludge. They were evaluated against thermochemical and biochemical treatment options. More specifically, those were (i) anaerobic digestion, (ii) incineration, (iii) gasification, (iv) slow pyrolysis and (v) hydrothermal carbonization (HTC). The proposed technologies were either considered individually or in combination and were supplemented by necessary upstream and downstream processes. To depict the entire life cycle, each treatment path started with the waste collection and ended with the generation of thermal and/or electrical energy and the lawful treatment of by- and co-products. The technological approaches in this work fall in the category Waste-to-energy (WtE) and can primarily be allocated to energy recovery from waste, the fourth step of the waste hierarchy. This approach may raise the question, why reuse and recycling, the initially preferable treatment steps for waste, were omitted. Reuse refers to "checking, cleaning or repairing recovery operations" (Article 3, 16 Directive 2008/98/EC) and seems counter-intuitive for organic waste. Recycling on the other hand appears meaningful and would in this context mainly refer to returning nutrients from the waste streams to the nutrient cycle. Within this work nutrient recovery is however always considered as a downstream option for biochemical treatment. Previous research proved that an energy exploitation step according to anaerobic digestion prior to the recycling of nutrients would always enhance the environmental performance in contrast to aerobic stabilization without energy recovery (LAURENT ET AL. 2014; KNAPPE ET AL. 2019a).

In combination with the respective treatment paths the sustainability aspects for each of the named substrates were evaluated. Social aspects were however not considered individually. As all management options fell in the same category that only differ in their technological approach, it was expected that aspects to *human well-being* such as public acceptance, space consumption or occupational health were of reduced interest. Additionally, it was assumed that adverse effects to *human health* were sufficiently described by the environmental analysis. Still, the lack of depicting social aspects would have to be considered a shortcoming of this study. Within the sustainability assessment of this work, the focus laid upon the environmental aspects. In alignment with the *Circular Economy Law* and the *European Waste Framework Directive*, it was also determined whether treatment paths were economically feasible. Economic aspects were only considered an exclusion criterion for a treatment path if the monetary costs were disproportionately increased.

In an abstracted form, the aim of this research can be described as "the coupling of an explicitly defined organic waste stream with the most environmental waste-to-energy process chain under the boundary condition of economic feasibility". To achieve this meta goal, the following subsidiary aims were pursued:

- Definition of the chemical composition that is representative of the respective waste stream. This comprised the dry matter (DM) content, the organic dry matter (oDM) content, the elemental composition and the heating value.
- The individual distribution of the energy, mass and elements of the substrate during the treatment steps.
- The energy consumption and production patterns within each process chain.
- The extension of energy and mass flows by emission data, ancillary and capital goods to compile a comprehensive environmental life cycle assessment (eLCA).
- The determination of investment, operation and maintenance and capital costs of treatment paths to determine the levelized costs of exergy (LCOE).

Based on this framework the following points were determined:

- the energy conversion efficiency for different treatment approaches,
- the environmental burdens resulting from different waste treatment paths and substrates,

- the marginal energy costs for the investigated treatment paths of the investigated substrates,
- whether source segregation of these waste streams is worthwhile, and to which extend it should be conducted,
- which technological setups are preferable from an environmental point of view, while economic burdens are not disproportionately elevated and, in this context,
- whether the implementation of alternative treatment technologies can lead to a decisive edge when treating organic waste streams.

The following questions cannot be answered in the realm of this work:

- the technological feasibility of treatment technologies in connection with the considered substrates,
- the comparison to treatment options that exclusively apply material recovery, as only
  options are considered that recover energy,
- social impacts resulting from the considered treatment paths.

#### The study is of high relevancy for:

- operators of waste treatment systems that aim at both economic and environmentally friendly options in the realm of municipal waste treatment,
- legislators, to align laws that foster sustainability to scientifically substantiated findings and to potentially promote promising technologies,
- the scientific community, to exploit datasets and modelling approaches for models on energy distribution, environmental assessment and economic computations.

Hereby the following boundary conditions applied. With regards to the temporal scope, the reference year was 2020 and the waste treatment systems were aligned to the legal regulations, which applied in this year. If possible, all background data were linked to this year. The geographical scope of this work was Germany, but the results may be applicable to countries with a similar waste composition and technological state-of-the-art.

To put the relevancy of this work into context, Chapter 2 gives an extensive literature review on the previously conducted research in the realm of environmental assessment of waste-to-energy technology. The current state of knowledge, research gaps and additionality of this work can be derived from this step.

	Chapter 2
Chapter 2	

Critical review on life cycle assessment of conventional and innovative waste-to-energy technologies DOI: 10.1016/j.scitotenv.2019.03.449

#### 2.1 Transfer of previous studies to this research

A transfer of previous research endeavors to the current research aim is essentially not possible. Several constraints hinder the direct application of previous results to the goal and scope as outlined in 1.4. The largest drawback results from methodological inconsistency between the studies. During the conduct of eLCA, practitioners are given many degrees of freedom with regards to methodological choices in the modelling phase. The studies differ by the extend of the system boundary, allocation rules, the choice in the functional unit (fU) or the LCIA methodology. To truly compare different technologies, a consistent methodology must be applied throughout all assessments. Furthermore, most assessments do not comply with the demands in a high quality eLCA. Thus, their validity is questionable. The geographical and temporal scope given in some of the studies would also exclude their application. Studies from countries with different waste compositions and a different technological state-of-the-art may not be applicable for Germany. Equally, studies that have been conducted several years ago would not represent the current situation. Regulatory policies have tightened, technological performances were enhanced and the degree of detail of eLCA databases has improved. Finally, the (to be) considered substrates were not assessed for all proposed treatment paths. Especially a cascading of treatment paths was rarely investigated or put into relation. Nevertheless, the previous studies can be harnessed with regards to their background datasets, namely the LCI. If available and suitable, they can be partly adapted to, and integrated into this research. Secondly, with respect to the process chain, a clear outline of necessary processes can be deduced from the previous research, especially, if product systems were based on actual industrial plants.

This work used a consistent methodological approach, which fulfilled the standardized criteria listed in the ISO 14040 and 14044 to conduct an eLCA. This implied that environmental compartments beyond the global warming potential were assessed and interpreted and that the system boundaries contained all relevant processes within a cradle-to-cradle/grave approach. Professional and comprehensive databases were utilized to model the background datasets. The foreground data was based on conducted analyses, real data from industrial plants, previously modelled literature datasets and individually modelled flows and processes. The modelled emission data were cross validated against the German legislation. Especially for crucial datasets (e.g., biomethane potential of substrates, heat demand of a HTC plant etc.) a multitude of these sources were utilized to guarantee robust data. These datasets were detailed and fully disclosed to allow for a high degree of transparency. Uncertainties were assessed for each individual flow and contracted in the realm of a Monte-Carlo simulation.

	Chapter 3
Chapter 3	

**Background on Substrates and Technologies** 

#### 3.1 Substrates

The four considered substrates do not cover all of the organic waste fractions, but represent the most dominant organic fractions of the municipal and commercial waste in Germany (DESTATIS 2020a). Minor shifts of current waste treatment systems towards more environmentally friendly technologies would thus result in proportionally high impacts. Additionally, the considered substrates cover a wide range of fuel characteristics (e.g. flowable sludge vs. solid wood, high ash vs. high organic content, highly biodegradable vs. complex biopolymers etc.) and recommendations for other biogenic material can be approximated from these findings.

#### 3.1.1 Waste wood

Waste wood is subdivided into five categories in Germany and must be collected separately if this is considered technically and economically feasible (Waste Wood Ordinance (AltholzV)). The category of a waste wood species is dependent on the level of contamination. A I refers to natural wood, while A IV refers to wood that was subject to a heavy application of wood preservatives. The fifth category is referred to as PCB-wood and contains wood species that were treated by means of polychlorinated biphenyls. The amount of treated waste wood totals to ~10 Mio. tons/a in Germany (FLAMME ET AL. 2019). Most of it resulted from the industry (2.55 Mio. tons) and from construction and demolition (4.04 Mio. tons). Two tons originated from the private sector and ~1 ton was imported. While ~80% of the waste wood could be allocated to A I and A II, only ~20% was classified as A III and A IV wood. For waste wood of classes A I and A II, an energetic use of the substrates competes with material recovery. Although the circular economy law propagates a cascaded utilization of waste wood for material recovery, in reality 75% is utilized for energy recovery. A subsidization under the Renewable Energy Sources Act (EEG) fostered the energetic exploitation in the past. A renunciation from the energetic exploitation from waste wood - which is principally eligible for material recovery - is forecasted, once the respective government funding will expire (UBA 2018). In this study only waste wood of the classes A III and A IV is considered. It represents hazardous waste and material recovery is considered little feasible (AltholzV). If a legal presumption is applied, the share of A III-wood amounts to ~1 Mio. tons/a and represents mainly bulky waste. The effective share of AIII waste wood in bulky waste is however considerably small (FLAMME ET AL. 2019). Waste wood of the class A IV mainly stems from construction and demolition activities and totals to ~1 Mio. tons/a.

#### 3.1.2 Food waste

As of now, food waste is not collected separately at a household level. Solely, businesses that process or sell animal products commercially, such as eateries or food producers, are required to collect the respective residues separately (~1 Mio t/a). Food waste includes all edible and non-edible parts, which are disposed of during any stage in the life cycle of a food product. A total potential of ~ 13 Mio. t/a is assumed in Germany, 50% of which is generated in households (SCHMIDT ET AL. 2019).

#### 3.1.3 The organic fraction of municipal solid waste

In the context of this work, the OFMSW comprises biowaste collected in biowaste bins (waste code: 20 03 01 04). Therein also household food waste is included. Since 2015 a source segregated collection of biowaste is mandatory in Germany and currently ~5 Mio t of OFMSW are collected per year (DESTATIS 2020a). A higher potential is given, due to poor separation efficiencies of the post-consumers and because several communities do still not provide separate waste bins. According to KNAPPE ET AL. (2019a) the unexploited potential for OFMSW is approximately 4-5 Mio tons, which is currently disposed of in residual waste bins.

#### 3.1.4 Sewage sludge

Annually, a total of 1.7 Mio t of sewage sludge (measured as dry matter) are disposed of from waste water treatment plants (WWTP) (DESTATIS 2018b). The amount of solid material entering the WWTP is higher because anaerobic digestion plants are often an inherent part of WWTP and convert a share of the solid material to sewage gas. Sewage sludge is a challenging substrate. It is rich in the finite resource phosphorous (P), but at the same time, raw sewage sludge holds a high ash and water content and is highly polluted with organic and inorganic contaminants. An amendment of the sewage sludge ordinance took effect in 2017 and soil related recovery options will be prohibited for large-scale WWTP from 2029 on, while P must be simultaneously recovered from sewage sludge.

#### 3.1.5 The role of organic waste in the energy sector

The current primary energy demand in Germany is >12,000 PJ/a and the final energy consumption is ~9,000 PJ/a (AGEB 2019). The (chemically bound) energy content of all considered organic waste fractions multiplied with their respective waste production totals to ~200 PJ/a. Additionally, most organic waste streams are characterized by a poor fuel quality and the conversion rate into useful (final) energy is low. This rough initial evaluation already shows that

WtE of organic waste residues cannot contribute considerably to the energy sector in the national scope. This raises the question, why such a high share of research is devoted to optimizing the WtE treatment paths for organic waste.

WtE-systems form the interface between waste management and energy generation. Similar to the KrWG, the declared objective of the energy sector in Germany is to arrive at a sustainable development (e.g., EEG). When comparing WtE-system to other energy systems they may not be able to compete with the environmental and economic performance of renewable energy systems such as wind, hydro and solar energy (WERNET ET AL. 2016; QUICKER ET AL. 2017). Given that the share of renewable energy sources increases in the future, WtE systems may eventually account for a disproportionately high share of the burdens in the energy mix. However, WtE-systems are and will remain a part of the waste management sector (QUICKER ET AL. 2017). They state a viable treatment option if reuse or recycling are not tenable or feasible. Hence the question, to which extent WtE-systems can contribute to the energy mix is not meaningful and it should not be the starting point of any discussion how WtE competes with other energy systems. WtE options must rather be contrasted against other WtE options or other waste management options in general. Research on this topic is therefore motivated by complying with the mindset of a circular economy, where a reduction of harmful impacts to the environment and human beings and the conservation of resources is the paramount objective.

#### 3.2 Treatment technologies

The considered treatment technologies in this work covered two proficient and well-established technologies in the German WtE sector: anaerobic digestion and incineration. Alongside these conventional technologies, three "alternative" technologies were considered: gasification, slow pyrolysis and HTC. They are promising options to treat waste fractions, but so far, they hold a negligible relevance in the waste management sector in Germany (UBA 2018). While both gasification and pyrolysis plants are available on the industrial scale since the 1970, they since failed an extensive market launch (GLEIS MARKUS 2012; UBA 2018). The HTC process on the other side is mostly only realized in lab- or pilot-scaled plants, but on the verge to be implemented at an industrial scale. An overview over the considered technologies is given in Appendix E. Currently, the ideal niche for advanced technologies has to be found. For mixed municipal waste, it is not expected that they outperform classic waste incinerators (QUICKER ET AL. 2017). The increasing degree of source segregation of waste streams and changes in the legislation (e.g., the amendment of the sewage sludge or the mandatory source segregation of OFMSW) bring these technologies however back into play.

	Chapter 4

# Chapter 4

**Materials and methods** 

#### 4.1 Considered treatment systems

The treatment of four organic waste species by five different principal conversion technologies were analyzed. These conversion technologies were either considered individually or in a cascade. The models were based on industrial-scaled plants and were linked to fixed throughputs:

- OFMSW: average throughput=40,000 t/a.
- Food waste: average throughput=10,000 t/a.
- Wood waste: average throughput=10,000 t/a.
- Sewage sludge: sludge resulting from the treatment of 600,000 person equivalents (PE). This is equal to ~9,500 t DM.

#### 4.1.1 Treatment paths

For each substrate six to eleven treatment paths were conceived and in total 35 treatment paths were formulated. On a meta-level the treatment paths can be summarized according to Figure 3. The boundary condition for all treatment paths was the incorporation of at least one conversion step of the substrate into heat and/or electricity. Simultaneously, it was not admissible that any paths ended with the provision of a secondary energy carrier (e.g., high calorific gas or char). They need to be further converted into heat and electricity. Agricultural application of digestate on the other hand was allowed for. All treatment paths, except those for sewage sludge, incorporated the initial waste collection, transport to the plant and transportation between treatment plants that were spatially separated. The latter applied, if secondary co-products were transported from the first treatment plant to a centralized waste incineration plant (T1) or a lignite power plant (T2). More information on transportation is given in Appendix F.

Treatment paths which contained an anaerobic digestion step were excluded for waste wood, since wood cannot be valorized during this process. For sewage sludge on the other side, it was assumed that anaerobic digestion was always included. Currently all 235 WWTP in Germany, exceeding a capacity of 100,000 person equivalents (PE), contain an anaerobic digestion stage (HABERKERN AND RETAMAL PUCHEU 2020). Consequently, this reduced the possibilities of potential treatment paths. While anaerobic digestion certainly states a proven and manageable technological approach to stabilize sewage sludge, it may rightfully be argued that anaerobic digestion may contradict a thermal post-treatment step. Sewage sludge states a substrate with poor fuel properties (high share of inert material, high moisture contents and an increased share of halides). An initial anaerobic digestion step decreases the organic matter content in the substrate and further deteriorates the fuel characteristics. The respective assessments were however beyond the scope of this work.

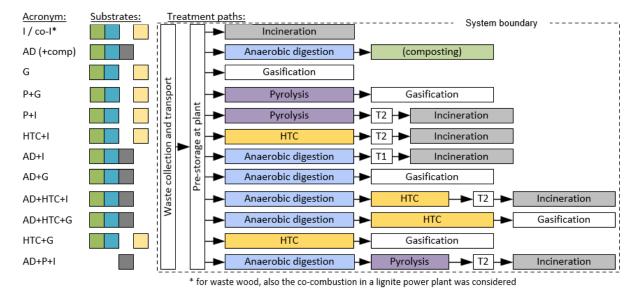


Figure 3: Considered treatment paths. T1=transport from first treatment plant to a waste incinerator, T2=transport from first treatment plant to a lignite power plant. For the substrates the following color codes apply: ■=OFMSW, ■=food waste, ■=sewage sludge, ■=waste wood

Pyrolysis of anaerobically digested material was excluded, as the HTC appeared to be a more suitable technological approach to carbonize wet substrates. One exception was the treatment of digested sewage sludge. Since all treatment steps for sewage sludge involved anaerobic digestion, also the pyrolysis of digested sewage sludge was investigated.

#### 4.1.2 Treatment modules

As seen in Figure 3, each treatment paths was composed of "treatment modules", which again contained several sub-processes that are detailed in Figure 4. Due to the different nature of each considered substrate, the modules had to be individually adapted to the treated substrate. For certain substrates or treatment paths sub-processes either needed to be incorporated or could be omitted (indicated by dashed frames). Since the different treatment required an individual adaptation depending on the treated substrate, a short description for each substrate and treatment path is given in Appendix H. Additionally, individual flow sheets that detail the mass and energy flows are given in the Electronic Annex. Subsequently a short overview over the model setup is given.

Module Anaerobic Digestion: During anaerobic digestion, substrate was digested at 37°C (mesophilic range). The biogas was combusted in a CHP-plant. For OFMSW, a dry fermentation according to the Valorga process (WOLF 2013) was implemented. Therefore, the substrate was mixed with water to reach a DM-content of 28 wt.-%. For food waste, wet fermentation was assumed and the DM-content in the reactor was set to 12 wt.-%. For sewage sludge on the contrary, the DM-content of the sludge in the feed stream needed to be increased prior to digestion. By means of a gravity thickener the initial DM-content was raised from 2 wt.-% to 4

wt.-%. Digestate from the fermentation processes was either spread on agricultural areas or further utilized for energetic recovery. For the latter case, a pre-drying step may be applicable.

If digestate was spread on agricultural areas, hygienization was required. For *OFMSW*, the digestate was separated into a solid and liquid part. The solid part was composted and the liquid part was thickened and pasteurized. For *food waste* and *sewage sludge*, the entire feed stream was pasteurized. If digestate was however utilized for an energetic recovery, a solid/liquid separation was implemented, to decrease the water content and to increase the lower heating value (LHV). The solid digestate did not require hygienization. Depending on the subsequent post-treatment, pre-drying was additionally included to further increase the LHV.

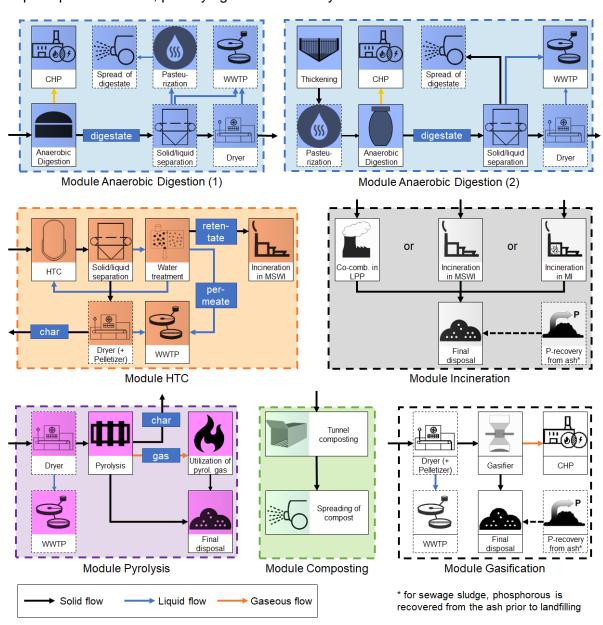


Figure 4: Sub-processes of each WtE-module. Dashed lines indicate processes that are optional depending on the treatment path and substrate. Module Anaerobic Digestion (1) refers to the substrates OFMSW and food waste, Module Anaerobic Digestion (2) refers to the treatment of sewage sludge. LPP=lignite power plant, MSWI=Municipal solid waste incinerator, MI=Mono-Incinerator.

Module Composting: This module only becomes relevant for OFMSW. Digestate from OFMSW underwent aerobic stabilization by means of tunnel composting within an enclosed building. Subsequently, the compost was spread on agricultural areas.

Module Incineration: Substrate was either co-combusted in a lignite power plant (char/wood), a waste incinerator or in case of sewage sludge in a mono-incineration plant (fluidized bed incinerator). Similar flue gas treatment systems and removal efficiencies were assumed. The solid residues were landfilled, while highly polluted filter ash was previously solidified by means of cement. For sewage sludge, P was recovered from the ash, prior to its final disposal. Therefore, a market mix (equal shares) of AshDec, Leachphos, EcoPhos, RecoPhos, PASCH and the Fertilizer Industry was created. This was considered the only option for P-recovery from sewage sludge, even though a greater number of concepts are available (e.g., direct recovery from the wastewater or sludge or recovery from pyrolysis or HTC-char).

Module HTC: The substrate was mixed with water until a biomass:water ratio=0.15 was achieved. The suspension was then sent to the HTC -reactor and carbonized at 200°C (220°C for waste wood) and for a retention time of 3 h to reach a lignite-like product. After the carbonization process the suspension was separated into a liquid and a solid fraction. The liquid fraction underwent a water treatment and was thereby purified by a cascade of nanofiltration and reverse osmosis. A share of the retentate and permeate were recirculated. The remainder was either incinerated in a municipal waste incinerator (retentate) or sent to a WWTP (permeate). The solid part (mechanically dewatered hydrochar) held a water content of 40 wt.-%. Hydrochar was either directly utilized within a thermal post-treatment step or it was dried (>85 wt-% DM) and pelletized for better transportability, storability, to avoid biological degradation and/or to enable gasification.

Module Gasification: For most substrates, the water content had to be reduced to allow for a gasification process and therefore drying became essential. Depending on its fuel characteristics, the substrate was gasified in a fluidized bed gasifier (material with a high share of fines, such as sewage sludge or dried OFMSW) or a fixed bed gasifier (bulk material, such as wood chips or pelletized char). The syngas was combusted in a CHP plant after it was purified and residual ash was landfilled. For sewage sludge, P was recovered from the ash in an identical way as presented for the module incineration.

Module Pyrolysis: Depending on its water content, substrate was pre-dried to a water content of ~20 wt.-%. Substrate was then pyrolyzed at 500°C (reactor temperature) for ~1h in a rotary kiln. The pyrochar was discharged from the kiln and the gaseous stream, consisting of pyrolysis gas and yet uncondensed pyrolysis oil, were jointly combusted in a combustion chamber.

A share of the produced heat was used to satisfy the parasitic heat demand of the rotary kiln. The remaining heat was used to drive a condensation turbine or to pre-dry the substrate.

#### 4.2 Waste composition

To conduct a thorough assessment of the present WtE paths, an implicit definition of the substrate material was required. Table 1 states the chemical composition of the considered substrates in this work. A more detailed breakdown is given in the Electronic Annex. Based on the composition, energy and mass distributions could be modelled, but also emission rates and nutrient recovery rates could be derived from the elemental composition.

Table 1: Key parameters and elemental composition of the considered waste streams (base case). FM=fresh matter, wt-%= weight per cent, wf=water free, I HV=lower heating value

Parameter/ Element	waste wood	r free, LHV=lower heati food waste	OFMSW	Dewatered sewage sludge (digestate) <sup>a</sup>
DM [wt%]	80.0%	25.0%	37.7%	25.0%
oDM [wt%, wf]	92.7%	93.5%	74.6%	60.0%
LHV [MJ/kg FM] <sup>b</sup>	14	3.8	4.2	2.2
C [wt%, wf]	46.91%	50.86%	38.25%	37.82%
H [wt%, wf]	5.74%	7.07%	4.56%	3.19%
N [wt%, wf]	0.49%	3.78%	1.55%	4.41%
O [wt%, wf]	39.50%	31.54%	30.94%	13.67%
S [wt%, wf]	0.01%	0.26%	0.16%	0.91%
P [wt%, wf]	0.01%	0.51%	0.24%	2.56%
K [wt%, wf]	0.07%	1.00%	1.01%	0.38%
CI [wt%, wf]	0.04%	0.96%	0.56%	1.10%
As [mg/kg]	5.6	0.4	2.5	68.0
Cd [mg/kg]	1.0	0.1	0.6	6.4
Co [mg/kg]	1.3	2.0	2.6	8.0
Cr [mg/kg]	100.0	3.7	16.0	145.3
Cu [mg/kg]	55.0	10.1	25.0	295.8
Hg [mg/kg]	0.11	0.02	0.09	3.00
Ni [mg/kg]	10.0	1.9	9.2	77.0
Pb [mg/kg]	14.0	0.7	44.0	184.0
Zn [mg/kg]	96.0	34.0	171.0	787.0
Fe [mg/kg]	785	215	5605	32470
Al [mg/kg]	1000	756	3860	16000
Hg [mg/kg]	0.11	0.02	0.09	3
Sources	FRITSCHE (2004) GRAS (2002) KUTSCHERA AND WINTER (2006) ECN.TNO (2020)	RIBER ET AL. (2009)	Mayer et al. (2020) Doka (2017)	ROSKOSCH AND HEIDECKE (2018) DWA (op. 2014) STATISTISCHES BUNDESAMT (2018) ECN.TNO (2020)

<sup>&</sup>lt;sup>a)</sup> the composition of raw sewage sludge was not relevant for the evaluation and only digested sludge underwent further post-treatment (see 4.1) <sup>b)</sup> calculated according to MICHEL (1938)

#### 4.3 Reference unit

Prior to the evaluation of the treatment paths a reference unit had to be chosen. Throughout all assessments, environmental and economic burdens were referenced to this unit. WtE-systems for organic waste fulfil two main useful functions: (i) they treat waste and (ii) they provide energy. This allowed for two evaluation approaches. The first approach falls in the category of an input-based approach. Treatment systems are assessed with regards to the treated waste material. Environmental impacts and monetary expenses are in this case referred to a certain amount of treated waste (e.g., 1 kg of OFMSW). An output-based approach on the other side links environmental and economic burdens to the provision of a certain amount of produced energy. Both choices are equally justifiable, but it implies a statement on how waste is comprehended. Within an input-based approach, waste is understood as a problematic matter and therein waste treatment systems function as a disposal system. Within an output-based approach, organic waste streams are considered as a resource and in the realm of WtE-plants this resource is converted into a useful product.

Although both approaches contain specific advantages and disadvantages (Appendix G), this work chose an *output-based approach*, as waste was primarily perceived as a resource and the sole treatment of waste was not considered a function. In alignment with the Waste Framework Directive a high conversion of the chemically bound energy of a waste stream into energy was considered preferential.

In this work all burdens were referenced to the amount of produced and exported energy (net energy output). Within the treatment paths, two forms of energy were considered (thermal and electrical energy) and had to be merged. They were contracted according to their exergetic value. Exergy describes the theoretical maximum share of energy in a system that can be converted into work. As electricity can be completely converted into work, its exergy factor was set to 1. The amount of the credited electricity was the produced electricity in the system minus the parasitic electricity demand in the foreground system (net electricity output). The electricity demand to produce ancillary or capital goods was herein not included.

The exergy value of thermal energy was derived via the Carnot-Efficiency. It is essentially a function of the exported temperature and the ambient temperature. Equation 1 describes the mathematical relationship. The higher the difference between the ambient temperature and the temperature level of the provided heat, the higher is the exergy factor of the thermal energy. A temperature level of  $\vartheta_a$ =10°C ( $T_a$ =283.15 K) was taken, as it represented the approximate average air temperature of the current normal period (DWD 2020).

$$EX_{heat} = 1 - \left(\frac{T_a\left[K\right]}{T_{heat}\left[K\right]}\right)$$
 Equation 1

 $EX_{heat}$ =Exergy factor of thermal energy,  $T_a$ =ambient temperature [K],  $T_{heat}$ = temperature level of the exported heat [K]

In the models it was assumed that heat was exported to residential buildings for space and drinking water heating via a district heating network. For this purpose, a standard load profile was developed for residential buildings. The calculation steps and boundary conditions for exporting heat are detailed in MAYER ET AL. (2020). In short, centralized incinerators were always connected to a district heating system, while decentralized plants could only provide heat to a district heating system via satellite plants. Secondly the exported heat had to contain a certain heat level ( $\vartheta_{\text{heat}} = \sim 100$ °C) and low-temperature heat from CHP plants alone did not fulfill this criterion. Finally, when heat was exported, the temperature level of the provided heat was set to an inlet temperature of  $\vartheta_{\text{heat}} = 70$ °C ( $T_a = 343.15$  K).

For sewage sludge, a sensitivity analysis with respect to the reference unit appeared necessary and therefore also an input-based approach was evaluated. Environmental and economic burdens were linked to the treatment of a certain amount of raw sewage sludge. This was done for two reasons: Firstly, the amount of exported energy from sewage sludge was deemed to be limited due to its poor fuel properties. Secondly, the systems were outlined in a way that P was always recovered from sewage sludge. Hence, *all* treatment systems for sewage sludge fall into the category of recycling.

#### 4.4 Exergetic evaluation

Since exergy was of central importance for the assessment, as it represented the common denominator of the balanced burdens, it needed to be accurately calculated. A poor illustration of the heat and electricity production and consumption rates would lead to erroneous results of both the environmental and economic assessment. For incineration and anaerobic digestion profound empirical values were available and therefore, the modelling work was reduced. For HTC, gasification and pyrolysis, each component had to be modelled individually. The modelling environment for the modules according to Figure 3 and Figure 4 are given in Appendix I (module incineration), Appendix J (module anaerobic digestion) Appendix K (module HTC) Appendix L (module pyrolysis) and Appendix M (module gasification). To determine the energy distributions within the treatment system, it was inevitable to calculate the mass and elemental distribution alongside. A conservation of mass, energy and elements had to be always maintained.

Produced and consumed electrical energy was implemented as a static value, as it was assumed that electricity could always be fed to and taken out of the national grid. For thermal energy it was assumed that production and consumption at the treatment site had to be aligned. Remaining heat had to be provided by the combustion of support fuel. Some components in the treatment systems were highly affected by weather conditions (e.g., heat demand for digester or heat demand in a district heating system). They were modelled dynamically (temporal resolution=1 h), to account for certain time points, when the heat demand exceeded the onsite heat production. In this case support fuel became necessary. In general, all systems were optimized to keep the share of external heat to a minimum. This meant that heat, which was provided on-site, was also utilized on-site, given that the temperature level of the generated heat matched the temperature level of the required heat. Only if additional heat was required, it was provided by an external heat source.

To rate the exergetic performance of the respective treatment paths, the "exergy:support fuel" ratio was introduced. It reflected a vector that contained the amount of the net exergy that was produced from 1 kg of fresh mass and contrasted it with support fuel, which was required to enhance the fuel properties of the substrate or its derivatives. Fuel required for transportation, the flue gas treatment or the production of capital or ancillary goods is therein not included. Initially a high net exergetic output suggested a high conversion rate of the chemically bound energy of the substrate. By combining this value with the share of support fuel, it could be determined, whether a high conversion rate was "purchased" at the expense of support fuel. This gave a more profound reflection of the exergetic performance.

#### 4.5 Environmental assessment

To determine the environmental burdens resulting from the different waste treatment paths, a consistent methodology had to be applied for all options. The methodology given in ISO 14040:2006 and ISO 14044:2006 was implemented within the eLCA-modelling framework. To maintain comparability, all modelling options that allow for degrees of freedom in this realm, were strictly defined.

#### 4.5.1 Goal and Scope

The goals of the eLCA were derived from the goals stated in 1.4. The eLCA aims at finding environmental sound WtE options for the four substrates under consideration, while disclosing the environmental mechanisms and unraveling environmental hotspots. The study was outlined for the year 2020 and the German scope. Technologies that have currently not yet reached market maturity are balanced according to an "ex ante" approach (Cucurachi et al.

2018). This means, they were modelled as fully functional, industrial-sized plants. The technological feasibility of the considered options could however not be guaranteed. Waste management treatment systems are complex systems that treat inhomogeneous material. The implementation of lab-scaled or pilot-scaled systems to continuously working, automated and industrial-scaled plants can be very lengthy or even fail in the long run.

#### a. Modelling environment

The software openLCA 1.9.0 (GREENDELTA 2020) was utilized to model and evaluate the product systems. The ecoinvent database 3.5 (WERNET ET AL. 2016) served as database for processes of the background system.

#### b. System boundary

All processes regarding the waste collection, intermediate transport, treatment and lawful disposal of by-products or spreading of useful co-products were considered. Intermediate waste storage at the post-consumer was however omitted. The system boundary was equal to that given in Figure 3 and is detailed for each treatment path in the electronic annex. Depending on the treatment path, the approach may be considered as a gate-to-cradle or gate-to-grave approach. If nutrients were returned to the nutrient cycle in the realm of digestate application to agricultural soils the approach may fall in the category gate-to-cradle. If treatment systems ended with the lawful disposal of byproducts, a gate-to-grave approach applied. The specific system boundaries of each system are given in the Electronic Annex.

#### c. Scope of the assessment

In the realm of the environmental assessment, both a comparative and hotspot analysis were prepared and discussed.

#### d. Functional unit

For all treatment systems the fU was 1 kWh of exergy. It was composed of the exergy of heat and electricity exported from a system. Heat was only credited if it was utilized beyond the system boundary. As outlined in 4.3, a sensitivity analysis with regards to the treatment of sewage sludge was conducted. Thereby the fU was set to 100 kg of raw sewage sludge (DM-content=2%).

#### e. Allocation

This assessment chose an attributional over a consequential approach. The inputs and outputs and lastly environmental burdens were linked to the fU by a normative rule. In comparison to a consequential LCA, the marginal changes resulting from either treatment system were not

considered. As the systems were subject to multi-functionality, allocation rules had to be defined. The first allocation rule referred to merging thermal and electrical energy by means of an exergetic approach (physical allocation). Further allocation was necessary when nutrients were recovered from the waste streams. According to the principle of allocation by system expansion, the shares of nitrogen, phosphorous and potassium were credited by offsetting them against mineral fertilizer (P as diammonium phosphate, N as calcium ammonium-nitrate, K as market for potassium fertilizer) and according to their mineral fertilizer equivalent (MFE).

When the fU was set to the treatment of 100 kg of raw sewage sludge, heat and electricity were no longer the fU and needed to be credited within the system boundary. Therefore, allocation by system expansion was chosen. Heat was offset against heat provision by natural gas (ecoinvent process: "market for heat, district or industrial, natural gas") and electricity was offset against the German electricity mix ("electricity, high voltage, production mix, DE"). The approach of allocation by system expansion is generally considered a trait of consequential (change oriented) LCA. In this case however, it must rather be viewed as an adjustment of the product system for an ecological equivalent (BRANDER AND WYLIE 2011).

#### 4.5.2 Life cycle inventory

The calculation of energy and mass balances formed the basic framework for the LCI. These data were complemented by data on emission factors, ancillary materials and capital goods. The general methodology to obtain these data consisted in an extensive literature research, the scanning of secondary databases (mainly WRATE (GOLDER ASSOCIATES 2014), GEMIS (IINAS 2020), Probas (UBA 2020) and BioEnergieDat (SCHEBEK ET AL. 2013)) and stoichiometric calculations. Furthermore, specific emissions were cross-validated against the current statutory regulations and, if necessary, adjusted. An overview over the main regulation with regards to waste treatment is given in Appendix N.

Where no data was available and could not be calculated or estimated through similar processes, data was omitted if it was unlikely to result in a relevant impact. Generally, the principle described in the ILCD handbook was followed: "Efficient and effective good practice in data collection requires to focus on what matters" (ILCD handbook 2010).

According to the ISO 14040, the LCI needs to be transparent and is therefore given in full detail for each product system and their processes. The LCI was prepared to a larger part in a parametrized form to retrace all calculations. Due to the complexity of the LCI, it is presented in the Electronic Annex.

## 4.5.3 Life cycle impact assessment

The ReCiPe 2016 Midpoint (H) methodology (HUIJBREGTS ET AL. 2017) was used to convert emissions and resource consumptions into environmental impacts. In total, 18 impact categories are comprised in this methodology. They are detailed in HUIJBREGTS ET AL. (2017).

## 4.5.4 Interpretation

Environmental impacts and their hotspots were critically revised and cross-validated against literature data to minimize errors in the resulting eLCA. To increase the robustness of the model, uncertainties were allocated to each flow (where applicable). Depending on the available datasets an adequate distribution type had to be chosen. Uncertainties resulting from poor data quality, e.g. data from lab-scaled results or data with little representativeness, were determined via the Pedigree-Matrix (CIROTH ET AL. 2016). The calculation basis for the Pedigree Matrix is given in Appendix O. The entire uncertainty of a product-system was computed numerically, by means of a Monte-Carlo simulation (1000 runs). To better analyze the multitude of impact categories, the ReCiPe 2016 Endpoint (H) methodology was used as an interpretation aid. Thereby, the environmental impacts of the midpoint categories are aggregated to three areas of protection, namely (i) damage to human health, (ii) damage to ecosystem diversity and (iii) damage to resource availability (HUIJBREGTS ET AL. 2017). Thereby the relevancy of the midpoint categories is put into context. Finally, the environmental impacts resulting from the production of electricity (German electricity mix) were extracted from the ecoinvent database to validate whether the modelled results were calculated in the right order of magnitude.

### 4.6 Economic assessment

For the economic assessment, the same fU was maintained as in the environmental assessment. Environmental burdens were linked to the production and export of exergy by means of *levelized costs of exergy* (LCOE). This represented a modified approach of the levelized costs of energy (KOST ET AL. 2018). Taxes were not included in the calculation approach and the LCOE must be understood as marginal net costs for exergy production from waste streams. Equation 2 defines this relationship. It should be noted that, even though it seems counterintuitive to divide a physical parameter (exergy) by an economic parameter (weighted average cost of capital), this is lastly only a result of several mathematical conversion steps (SALVADORES AND KEPPLER 2010).

$$LCOE = DC \cdot \frac{\sum_{t=0}^{n} \frac{I_{t} + (O+M)_{t}}{(1 + r_{DC} - i)^{t}}}{\sum_{t=0}^{n} \frac{exergy}{(1 + r_{DC} - i)^{t}}} + EC \cdot \frac{\sum_{t=0}^{n} \frac{I_{t} + (O+M)_{t}}{(1 + r_{EC} - i)^{t}}}{\sum_{t=0}^{n} \frac{exergy}{(1 + r_{EC} - i)^{t}}} = \cdots$$

$$= \frac{\sum_{t=0}^{n} \frac{I_{t} + (O+M)_{t}}{WACC^{t}}}{\sum_{t=0}^{n} \frac{exergy}{WACC^{t}}}$$
 Equation 2

DC/EC=share of debt capital and equity capital [ ], n=economic lifetime of the plant,  $I_{\leftarrow}$ Investment costs in the year t [ $\in$ /a],  $(O+M)_{\leftarrow}$ Operation and maintenance costs in the year t [ $\in$ /a],  $r_{DC/EC}$ =interest rate debt capital/equity capital<sup>1</sup> [ ], i=rate of inflation [ ], exergy=amount of exported exergy [kWh/a], WACC=weighted average cost of capital [ ]

For the case of sewage sludge, the economic burdens with regards to an input-based fU were additionally calculated. Equation 2 was adjusted to calculate the net treatment costs for 100 kg of raw sewage sludge. Equation 3 gives the calculation steps.

n=economic lifetime of the plant,  $I_t$ =Investment costs in the year t [ $\in$ /a],  $(O+M)_t$ =Operation and maintenance costs in the year t [ $\in$ /a],  $\dot{Q}_{el/th}$ =amount of exported electricity/heat [kWh/a],  $p_{el/ng}$ =net (industrial) price for electricity and natural gas [ $\in$ /kWh] WACC=weighted average cost of capital [],  $\dot{m}_{sew}$ =mass of treated raw sewage sludge (DM=2%) [kg/a]

Within Equation 2 and Equation 3, investment costs (I<sub>t</sub>) contained the costs for construction and equipment, including buildings, infrastructure, additional costs for engineering and authorization and land development that were purchased at t=0. Further investment costs included fees and contingencies. If one component required a replacement after its economic lifetime, the annualized costs were added to the project costs. If components exceed the lifetime of the plant, the discounted residual value was credited. Operation and maintenance (O+M) costs included costs for maintenance, working materials, insurance, fuel, transporting, personnel and overhead costs. Revenues for fertilizer were herein also credited. Substrate costs were not

<sup>&</sup>lt;sup>1</sup> Corporate tax rate for debt capital was not taken into consideration.

accounted for, since waste was considered free of charge. Like in the environmental assessment, waste collection and transport to the initial treatment plant were within the system boundary. Both investment and O+M costs were sourced from an extensive evaluation of data from literature. Cost structures were mainly taken from the German scope and were extended by studies from the European scope if data was scarce (e.g., for innovative technologies).

Cost data of different plant sizes were scaled up/down by means of a degression exponent. Equation 4 gives the relationship. The capacity of plants was herein measured as throughput. In few cases, when the throughput was not given, plants were scaled up according to their rated power. A default degression exponent of 0.6 was applied (COULSON ET AL. 1964), if no other scaling factor was presented in the literature data. If multiple trains were used, no degression was assumed (deg=1). Decentralized plants were scaled to their referenced throughput (as given in 4.1). Centralized plants, in which the referenced waste-streams were cotreated, were given a fixed throughput and the share of waste or waste derivatives were referenced without scaling. The lignite power plant was set to a throughput of 3.15E6 t char (90% DM)/a, the municipal waste incinerator to 240,000 t FM/a and the mono-incineration plant for sewage sludge to 40,000 t DM/a (average throughputs of plants located in Germany (UBA 2018)).

$$C_{ref} = \left(\frac{cap_{ref}}{cap_{lit}}\right)^{deg} \cdot C_{lit}$$
 Equation 4

 $C_{ref}$ =costs of the referenced plant, cap<sub>ref</sub>=capacity of the referenced plant, cap<sub>lit</sub>=capacity of the plant as found in the literature,  $C_{lit}$ =costs of the plant as found in the literature, deg=degression coefficient

As the design of the plants may vary greatly within each considered technology, a high variance was expected. The range of variation was accounted for by implementing a Monte-Carlo simulation (1000 runs) and the costs were varied within the respective bounds and according to their distribution. The implemented datasets for the calculation of the LCOE and treatment costs are given in Appendix P.

# 4.7 Application of the methodological framework

In Figure 5 the methodological framework is summarized. In the context of this cumulative work, it was applied stepwise to the considered substrates and technologies.

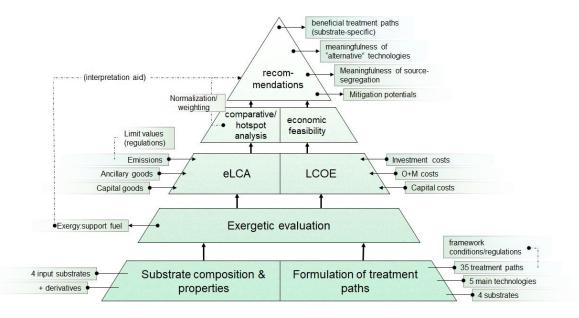


Figure 5: Summary of the methodological framework

Within Chapter 5, the described methodological approaches to quantify environmental and economic aspects were applied to OFMSW. Thereby the substrate composition and the biomethane potential of OFMSW were initially evaluated. Based on this input data, an eLCA and LCOE were performed for the treatment of OFMSW by means of anaerobic digestion, incineration and a combination of both.

Chapter 6 presents a study in which the scope of the substrates was extended from OFMSW to food waste and waste wood. The principal technologies were HTC, anaerobic digestion and incineration. They were considered individually or in a cascade.

In Chapter 7 the post-treatment of sewage sludge according to direct spreading and monoincineration were compared from an environmental point of view. Additionally, it was tested whether an intermediate carbonization step by means of HTC or pyrolysis was beneficial.

Finally, in Chapter 8, the resulting environmental and economic burdens for all presented treatment paths according to Figure 3 were compiled, discussed and conclusions were drawn accordingly.

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# **Chapter 5**

Economic and environmental life cycle assessment of organic waste treatment by means of incineration and biogasification. Is source segregation of biowaste justified in Germany?

DOI: 10.1016/j.scitotenv.2020.137731

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# **Chapter 6**

Life cycle assessment on the treatment of organic waste streams by anaerobic digestion, hydrothermal carbonization and incineration DOI: 10.1016/j.wasman.2021.05.019

	Chapter 7
Chapter 7	

Life cycle assessment of prospective sewage sludge treatment paths in Germany

DOI: 10.1016/j.jenvman.2021.112557

Chapter 8
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# **Chapter 8**

**Results and Discussion** 

Conclusively, all considered substrates and treatment paths were analyzed. Treatment paths, which were previously not addressed (Chapter 5-Chapter 7), are now fully implemented in the evaluation. In the following, the respective treatment paths for the four considered substrates are discussed in an abridged version. The analysis consisted of three compounds, (i) the exergetic performance, (ii) the economic analysis and the (iii) environmental assessment.<sup>2</sup>

#### 8.1 Waste wood

### 8.1.1 Exergetic performance

Initially the exergetic performance was evaluated according to the "exergy:support fuel" ratio. The evaluation is displayed in Figure 6. This vector does not state an assessment criterion itself but is strongly linked to the environmental and economic evaluation and serves as interpretation aid. A high specific exergy output and a low demand in support fuel were hereby preferable.

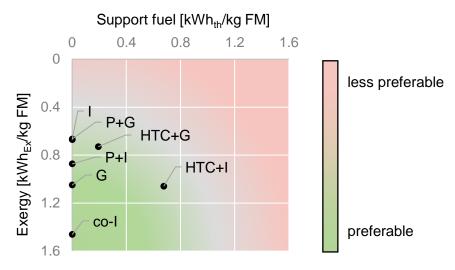


Figure 6: Exergy:support fuel ratio for the considered treatment paths for waste wood.

*Incineration:* The highest fuel utilization was given for the co-combustion of waste wood ("co-I"). Waste wood was converted at an efficiency of 38% into electricity (with respect to the LHV) and the specific exergy output was ~1.5 kWh<sub>Ex</sub>/ kg waste wood (measured as FM). Municipal waste incinerators showed lower electric efficiencies, but heat was co-produced alongside. As

<sup>&</sup>lt;sup>2</sup> It should be noted that the presented results of both the environmental and economic assessment may differ from those given in Chapter 5-Chapter 7 as the methodology was customized in the realm of the guidelines of the respective journals and adjusted throughout the reviewing process. Additionally, the datasets were further developed and adjustments were conducted in order to incorporate new insights (e.g. implementation of lab data). The core statements did however not change.

the exergy content of exported heat is only  $EX_{heat}=\sim0.17$  and because only  $\sim50\%$  of the produced heat could be utilized by the end user (unused heat during summer and heat losses in the district heating system), the specific exergy output decreased to  $\sim0.7$ kWh<sub>Ex</sub>/kg FM. Support fuel was not required in either case.

Gasification: A conversion by means of gasification ("G") was preferable, as waste wood was converted into exergy at a higher efficiency in comparison to waste incineration ("I"). Cold gas efficiencies were as high as 78% and the gas conversion efficiencies in a CHP plant were 37% and 53% for electricity and heat, respectively. As pre-drying was not required for waste-wood, no support fuel was inventoried, and heat could be exported to a district heating system. The specific output was 1.05 kWh<sub>Ex</sub>/kg FM.

*Pyrolysis:* During the pyrolysis of waste wood volatile gases were directly incinerated. The produced heat was partly deterred to maintain the heat level in the pyrolysis kiln and the remainder was utilized in a steam turbine. Steam parameters were similar as in a waste incinerator, but the turbine was designed in a way to produce electricity only. During the post-treatment of the pyrochar (co-combustion in a lignite power plant and gasification), the energy content of the pyrochar was further exploited at a high efficiency. The specific exergy outputs were 0.7 ("P+G") and 0.9 ("P+I") kWh<sub>Ex</sub>/kg.

*HTC:* During HTC of waste wood larger shares (~0.40 kg<sub>DM</sub>/kg<sub>DM</sub> input) were transferred to the liquid or gaseous phase and thus evaded an energetic utilization. Consequently, the overall fuel utilization rate was diminished. Specific exergy outputs were 1.0 and 0.55 kWh<sub>Ex</sub>/kg for "HTC+I" and "HTC+G". At the same time, support fuel was required for the HTC process and the post-drying step (in total ~0.7 kWh<sub>th</sub>/kg FM). In "HTC+G" the amount of support fuel could be reduced in comparison to "HTC+I", as high temperature heat could be utilized from the gasification module or more precisely the combustion of syngas in a CHP plant.

# 8.1.2 Environmental assessment

The LCIA by means of the ReCiPe Midpoint (H) method resulted in 18 impact categories. The relevance of these categories with regards to the treatment of waste wood was initially validated by weighting them according to the ReCiPe Endpoint (H) methodology. (i) Climate change, (ii) fossil depletion, (iii) human toxicity, (iv) particulate matter, (v) ozone formation and (vi) terrestrial acidification were found to have a major impact on the total environmental damage. As weighting is strictly subjective, impact categories were individually analyzed. It was found that other impact categories were of reduced interest, because impacts arose mainly in the background system or amounted to an insignificant impact (e.g. one order of magnitude smaller in comparison to impacts arising from the German electricity mix). The weighted results

are given in Figure 7. The lowest environmental burdens were indicated for "I", "co-I" and "G". As the endpoint categories may not stand for themselves, an in-depth analysis was conducted that incorporated an analysis of both the comparative and hotspot analysis. The LCIA evaluation of the main midpoint categories is given in Figure 8. The ranges of uncertainty are depicted as boxplots. The hotspot analysis is presented as circular bar plot in Figure 9. Both the comparative and hotspot analysis were considered jointly.

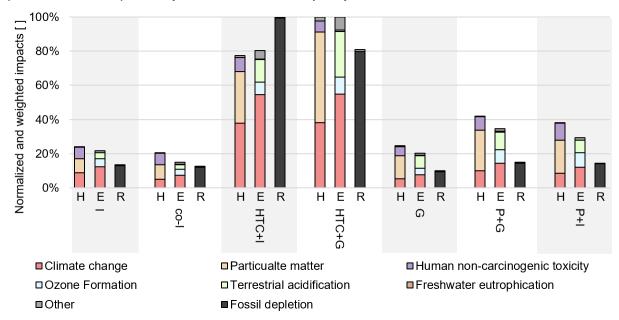


Figure 7: Normalized and weighted results for the considered treatment paths for waste wood according to ReCiPe Endpoint (H). H=Damage to human health, E=Damage to ecosystem diversity, R=Damage to resource availability.

#### a. Climate change and fossil depletion

Climate change and fossil depletion were strongly correlated in this case and may be discussed together. For treatment paths, featuring an HTC process, the utilization of a fossil support fuel (i.e. natural gas) dominated both categories (Figure 9). The total GWP and fossil depletion potential could be reduced by up to 65% if a renewable energy carrier was utilized. Heat provided by natural gas results in 0.25 kg CO<sub>2,Eq</sub>/kWh<sub>el</sub> and 0.1 kg oil<sub>Eq</sub>/kWh<sub>th</sub>, while heat from wood chips for instance accounts for only 0.02 kg CO<sub>2,Eq</sub>/kWh<sub>el</sub> and 0.003 kg oil<sub>Eq</sub>/kWh<sub>th</sub> (WERNET ET AL. 2016). Overall, HTC of waste wood did however not seem a plausible approach, as raw waste wood already holds good fuel properties and during HTC the LHV of the solid product only increased marginally. Additionally, ~40% of the input material were dissolved in the liquid phase (process water) or transferred to the gaseous phase (process gas). Lastly, the HTC-char had to be (i) dewatered and (ii) dried and (iii) the process water needed to undergo a post-treatment. As seen in Figure 9, these post-treatment steps accounted for larger impacts in both categories.

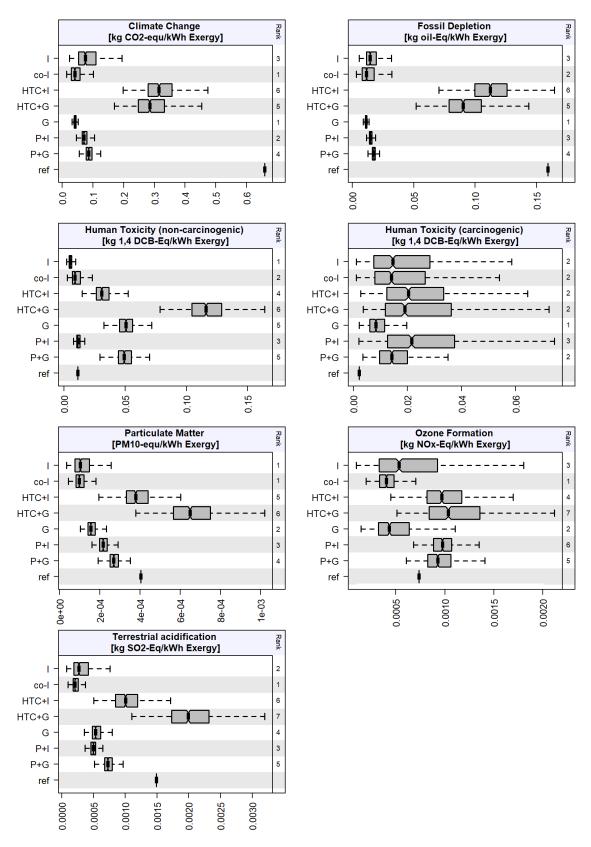


Figure 8: LCIA of the most relevant impact categories for the treatment of waste wood. The environmental impacts are presented as notched boxplots, whereby the uncertainty ranges were derived from the Monte-Carlo simulation. If notches did not overlap, it was implied that the environmental impacts differ significantly. A rank was allocated accordingly (1=lowest impact). The system "ref" accounts for environmental impacts associated with the production of 1 kWh of electricity in the realm of the German electricity mix.

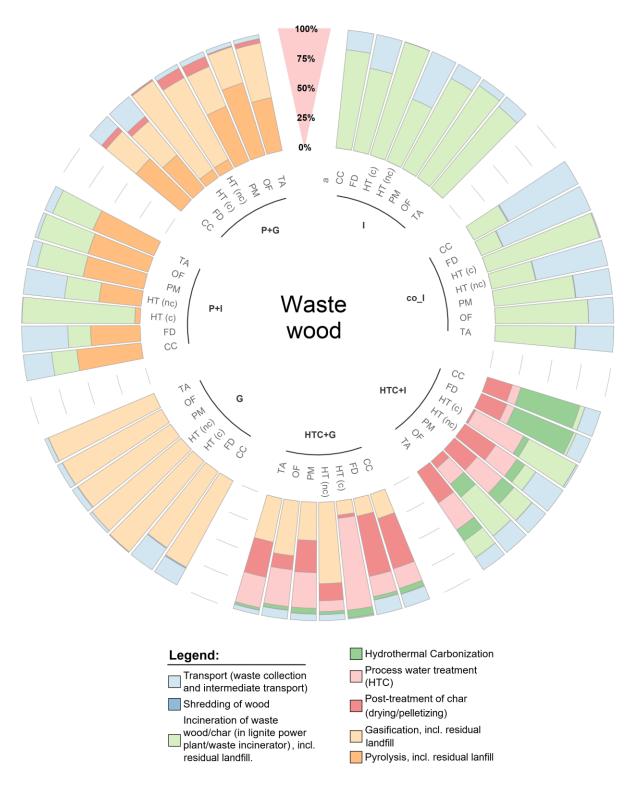


Figure 9: Hotspot analysis of the investigated treatment paths for waste wood. CC=climate change, FD=fos-sil depletion, HT (nc)=Human toxicity non-carcinogenic, PM=particulate matter, OF=ozone formation, TA=terrestrial acidification, FE=freshwater eutrophication. Results were normalized to 100%.

For all other treatment paths, no support fuel was required. In "I" however, a consumption of natural gas was implemented for preheating the flue-gas prior to the DeNox stage. This step is typically only required for low dust selective catalytic reduction (SCR), but the plant was

modelled according to an average waste incinerator (DOKA 2017). Otherwise, the fossil depletion and GWP resulted mainly from the utilization of ancillary and capital goods along the process chain and the initial transportation step. Cascaded treatment systems such as "P+G" or "P+I" entailed longer process chains and therefore a larger infrastructure. The impact of transportation was particularly large, where wood or its derivatives were transported to a lignite power plant (i.e. "co-I", "HTC+I", "P+I").

The direct emission of climate-changing gases was marginal. During the gasification of waste wood methane slips of ~1 vol-% may occur at the CHP plant. N₂O may be released during incineration in a waste incinerator and the pyrolysis process at low rates of ~40-60 mg/kg wood waste. Still, N₂O emissions accounted for ~20% (systems "P+I" and "P+G") – ~35% (system "I") of the total GWP due to their high characterization factor. As N₂O emissions are also highly relevant for other substrates, they shall be discussed in detail already at this point. In general, the share of fuel-N correlates with the N2O emission levels and is therefore relatively low for waste wood. Three main pathways become relevant for the N<sub>2</sub>O formation during thermochemical treatment. (i) Sub-stoichiometric oxygen conditions lead to a formation of N2O from fuel-N (e.g. in the primary combustion zone), (ii) a combustion temperature <850°C provokes an increased conversion of fuel-N to N<sub>2</sub>O and (iii) during the selective non-catalytic reduction (SNCR) N<sub>2</sub>O may be formed during the reduction of NO<sub>x</sub> (NEUWAHL ET AL. 2019). As waste incinerators guarantee combustion temperatures >850°C and a high excess air ratio in the secondary combustion chamber ( $\lambda=-1.4-1.8$ ), the main entry paths would be through the SNCR. Particularly, if urea is used as reactant the N₂O emissions increase from 1-2 mg/Nm³ flue gas to 20-60 mg/Nm³ (values refer to average waste, NEUWAHL ET AL. 2019). According to expert judgment, the N₂O emissions may on average be assumed to be 7 mg/Nm³ flue gas or 50 g/t MSW (CALVO BUENDIA ET AL. 2019). In the pyrolysis process an oxygen deprived atmosphere within the pyrolysis reactor is given and would increase N<sub>2</sub>O emissions. Most of the N<sub>2</sub>O emissions would however be oxidized to N<sub>2</sub> and O<sub>2</sub> in the over-stoichiometric burnout zone again. Regardless, the implemented data showed considerable N<sub>2</sub>O emissions (~17 mg/Nm<sup>3</sup> flue gas, average waste). They were sourced from a pyrolysis plant, where urea was utilized during the SNCR and it is hypothesized that this was the main entry path for N<sub>2</sub>O emissions. For gasification, no indications were found that substantial amounts of N<sub>2</sub>O were released with the flue gas. Accordingly low emissions were quantified. Also databases such as WRATE (GOLDER ASSOCIATES 2014), GEMIS (IINAS 2020) or BioEnergieDat (SCHEBEK ET AL. 2013) quantified negligible amounts of N<sub>2</sub>O during gasification processes. Sub-stoichiometric conditions during the gasification process would suggest that N2O is formed during the gasification process but would be oxidized again during the syngas combustion. The DeNOx stage after

the syngas combustion is usually designed as selective catalytic reduction (SCR) and would therefore exclude a re-formation of N<sub>2</sub>O. As N<sub>2</sub>O emissions depend largely on the operation mode, their factual emission is subject to a high uncertainty.

### b. Human toxicity

Toxic organic compounds were destroyed in the realm of a thermal treatment and were not deemed to play an important role in this context. Toxicity was mainly provoked by the release of heavy metals to the environment. As all thermal treatment plants were characterized by similar flue gas cleaning efficiencies, the direct emissions of heavy metals were similarly low per kg of treated waste wood.

Impacts in the category *carcinogenic human toxicity* resulted exclusively from the release of Chromium VI to the surface water, during the landfilling of the residues (slag and ash). Between lignite power plants and incineration plants slightly different transfer coefficients for Chromium VI were present, which led to slightly different impacts within both technologies. Chromium VI is a potent carcinogen, but its transfer from a landfill site to the surface water is subject to a high level of uncertainty (Figure 8).

Non-carcinogenic human toxicity was mainly associated with the production of ancillary goods, which were required for the treatment systems and the release of heavy metals during transportation (Figure 9). In systems that contained a gasification step a higher impact was calculated in this category. It stemmed from the production of methyl ester, which was implemented in gasification plants for tar removal from the flue gas and more precisely from growing rape. The application of fertilizer to rape, which is connected to the entry of heavy metals to the soil resulted in a major toxicity potential (mainly caused by the elementary flow zinc). This impact is however little justified as the amount of released heavy metals during fertilization is typically below a critical threshold value.

#### c. Particulate matter formation, ozone formation and terrestrial acidification

Particulate matter was released as such (primary particulate matter) and at a much larger rate as secondary particulate matter. NO<sub>x</sub> emissions were the main driver of the latter and resulted mainly from the thermal treatment of waste wood. As the treatment systems were outlined towards the German limit values stated in the 17. BimSchV, the specific emissions were similar per m³ of released flue gas throughout the different technologies. One exception was the co-combustion of waste in lignite power plants, where higher particulate matter emissions (measured as total dust) were admissible.

The impacts in the category particular matter formation were overall strongly correlated with the specific exergy output (Figure 6). A high exergy output reduced the specific flue gas amounts, simply because less material was combusted per kWh<sub>Ex</sub>. Additionally, (i) the combustion of support fuel, (ii) transportation activities and (iii) the consumption of ancillary and capital goods added to the particulate matter formation potential due to the release of NO<sub>x</sub> (Figure 9). Other emissions that are known to result in increased impacts in this category were SO<sub>x</sub> and NH<sub>3</sub> but were in this case released at a low rate.

Like particulate matter, *terrestrial acidification* was driven by NO<sub>x</sub>, SO<sub>x</sub> and NH<sub>3</sub> emissions. As the characterization factors are slightly different in both categories a marginally different pattern is seen for the two impact categories. Low level *ozone formation* (photochemical smog) mainly resulted from NO<sub>x</sub> emissions and to a much smaller extent on non-methane volatile organic compounds (NMVOC).

Throughout all of these three categories, the systems "I", "co-I" and "G" showed the lowest environmental burdens (Figure 8), due to (i) a high fuel utilization rate, (ii) no demand for support fuel and (iii) short process chains. The latter reduced the amount of ancillary and capital goods.

#### 8.1.3 Economic assessment

Figure 10 details the mean waste treatment cost and incorporates uncertainty ranges in the form of a violin plot. Waste was considered free of charge, but expenses for waste collection were accounted for. For all treatment options, this initial waste collection (assumption: waste collection at the place of generation) had an impact of ~10-30% of the total LCOE. In this context it should be noted that the data basis for waste collection was poor and may not reflect actual conditions accurately (Appendix P).

Gasification ("G"), waste incineration ("I") and co-incineration in a lignite power plant ("co-I") resulted in the most economic options. The mean expenses for treatment and waste collection were found to be <20 ct/kWh<sub>Ex</sub>. Hereby it may be assumed that the LCOE for gasification are to be found at the upper end of the uncertainty range, as a comprehensive flue gas system becomes necessary, when treating waste wood. Treatment options featuring a carbonization step ("HTC+I", "HTC+G", "P+G" and "P+I") were on the contrary the costliest option. On average they ranged between 30-45 ct/kWh<sub>Ex</sub>.

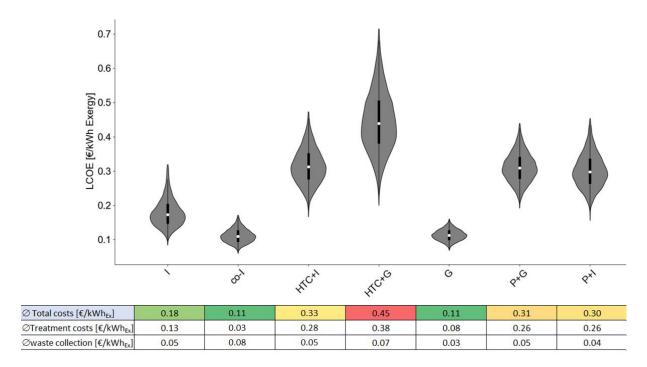


Figure 10: LCOE for different treatment options of waste wood. Violin plots represent the distribution curve for the total costs. Outliers were excluded (Z-score<3). Average total costs are separately given for the waste treatment itself and the initial waste collection (incl. transport to the first treatment plant).

#### 8.1.4 Conclusive recommendations

Based on the results of the economic and environmental assessment it was found that the implementation of a carbonization step of waste wood did not result in an added value. This excluded the treatment paths "HTC+I", "HTC+G", "P+I" and "P+G". On the contrary the cocombustion of waste wood in a lignite power plant ("co-I") represented the most beneficial option. In this case, it is necessary that the lignite power plant fulfills the criteria to co-combust hazardous waste according to the 17. BlmSchV in connection with the 4. BlmSchV. Additionally, the availability of a lignite power plant within local proximity to the waste collection site should be given. Transporting distances were modelled according to the average distance between any site in Germany and the closest lignite power plant (~200 km). Particularly in the southern part of Germany, this option may not be viable anymore as the transporting distances become unduly long. Gasification of waste wood in a gasifier (G) then appears to be a more sustainable solution. Similar economic impacts and marginally higher environmental impacts were found for this option. However market-readiness and the technological performance are crucial components that could reduce the attractiveness of this option. The modelled conversion efficiencies seem technically feasible (KALTSCHMITT ET AL. 2016), but are at this point slightly lower in reality (PATUZZI ET AL. 2021; SCHÜßLER ET AL. 2009). Secondly, gasification states a more complex and currently less resilient technology in comparison to incineration. Insufficient temperature stability in the reactor, substrate blockage or caking may run contrary

to a steady operation (QUICKER ET AL. 2017). The mass burning of wood waste in municipal waste incinerators resulted in a similarly high environmental performance in comparison to gasification. The economic expenses were higher, due to a poor conversion rate into electricity. If neither gasification nor co-combustion in a power plant are possible, this path is clearly recommended. Waste incineration represents a sturdy technology, with a high market availability.

In Germany, waste wood is currently mainly treated in wood mono-incineration plants that operate in a cogeneration mode. This option was not considered, but it is plausible that their environmental impacts are similar to those of a waste incinerator (option "I"), due to similar flue gas treatment systems and only marginally higher conversion efficiencies. Within these mono-incineration plants, waste wood of the categories AIII and AIV is currently always co-combusted with waste wood of the categories AI and AII (UBA 2018) to allow for higher throughputs and to reduce the efforts for flue gas cleaning.

## 8.2 Food waste

## 8.2.1 Exergetic performance

In Figure 11 the exergy:support fuel ratio for the considered treatment paths for food waste are displayed.

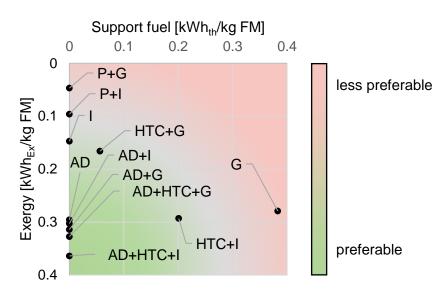


Figure 11: Exergy:support fuel ratio for the considered treatment paths for food waste.

Anaerobic digestion: A high utilization of the chemically bound energy of food waste was achieved for treatment paths that incorporated anaerobic digestion. For the respective treatment paths ("AD", "AD+I", "AD+G", "AD+HTC+G" and "AD+HTC+I") ≥0.3 kWh<sub>Ex</sub>/kg food waste (measured as FM) could be achieved. Food waste represents a substrate with a high water content (~75 wt.-%) and a high share in organic matter (oDM>90%), while complex organic

material, such as lignocellulosic material is hardly present. This makes it an ideal substrate for anaerobic digestion. During the decomposition both high biogas yields and methane contents were determined. More than 60% of the chemically bound energy of food waste were exploited by anaerobic digestion. A thermal post-treatment step of the digestate could only slightly increase the specific exergy output. This was mainly due to three reasons. Firstly, during the solid-liquid separation step, a significant share (~55%) of the chemically bound energy of the digestate was transferred to the liquid digestate and evaded an energetic utilization. Only the remainder - the solid digestate – was available for the thermochemical post-treatment. Secondly, little energy could be harvested from the solid digestate due to its poor fuel properties. Thirdly, the heat, which was previously exported to a district heating system (and thus increased the exergy output) was used for drying processes and/or the HTC process. The positive aspect of these implications was, that sufficient heat could be provided within the systems (i.e. "AD+I", "AD+G", "AD+HTC+I", "AD+HTC+G") to run autarkic with regards to their energy demand. In Figure 12 the described effects are exemplary illustrated for the two systems "AD" and "AD+I" on the basis of Sankey Diagrams.

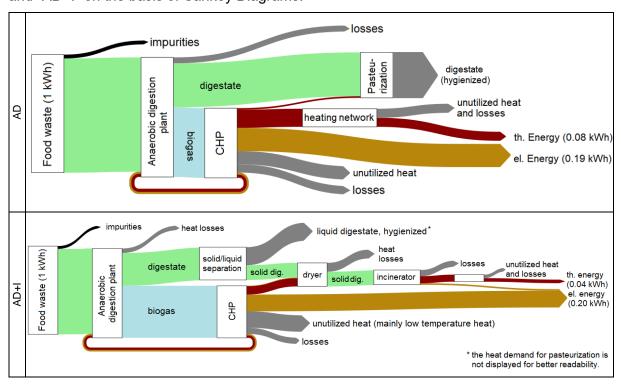


Figure 12: Sankey diagrams for the treatment paths "AD" (top) and "AD+I" (bottom). Thickness of lines refers to energetic content of flows. Chemically bound energy is displayed as LHV.

Incineration/Gasification: Direct thermo-chemical treatment of food waste was hindered, as larger shares of energy were needed to evaporate the chemically bound water from the raw substrate. In "I" this resulted in an overall decreased conversion efficiency of ~0.15 kWh<sub>Ex</sub>/kg FM. In "G" the effect could be counteracted by pre-drying the substrate with the aid of support

fuel. As a result, the amount of support fuel ( $\sim$ 0.4 kWh<sub>th</sub>/FM) exceeded the exergy output of the system ( $\sim$ 0.3 kWh<sub>Ex</sub>/kg FM).

*HTC*: Intuitively, also HTC appears to be a promising approach, as this technology is predestined for wet substrates. However larger shares of food waste were transferred to the liquid and gaseous phase during HTC. The utilizable product yield was only ~0.5 kg char<sub>DM</sub>/kg biomass<sub>DM</sub>. As a result, the exergy exploitation in "HTC+I" or "HTC+G" was reduced in comparison to the system "AD". A high share of proteins and the lack of complex biopolymers are known to reduce the transfer of biomass to biochar in this context (BLÖHSE 2017). Additionally, larger amounts of support fuel were required. The system "HTC+I" produced ~0.3 kWh<sub>Ex</sub>/kg FM and required ~0.2 kWh<sub>th</sub> of support fuel per kg FM. For "HTC+G" the amount of support fuel could be diminished (~0.05 kWh<sub>th</sub>/kg FM), as high temperature heat from the gasification process was available. This in turn also reduced the exergy output to ~0.15 kWh<sub>Ex</sub>/kg FM.

*Pyrolysis:* Within "P+G" and "P+I" the heat which was produced from the pyrolysis process was entirely consumed by the drying unit. A net energy output could only be achieved by the secondary post-treatment according to incineration or gasification. The systems ran autarkic, but the overall exergy yields were <0.1 kWh<sub>Ex</sub>/kg FM.

#### 8.2.2 Environmental assessment

An individual analysis and the weighting step could narrow down the LCIA to relevant impact categories. The impact categories (i) global warming potential, (ii) fossil depletion, (iii) human toxicity (non-carcinogenic), (iv) particulate matter formation, (v) ozone formation, (vi) terrestrial acidification and (vii) freshwater eutrophication were found to be of increased importance. The weighted results are given in Figure 13.

Like the exergetic evaluation, product systems that contained an anaerobic digestion step ("AD", "AD+G", "AD+I", "AD+HTC+I" and "AD+HTC+G") represented preferential options according to the weighted results. Direct thermochemical conversion according to "I" and "G" showed slightly higher environmental burdens throughout most categories. However, the fossil depletion potential was strongly increased. Treatment steps that contained an HTC process showed among the highest environmental burdens.

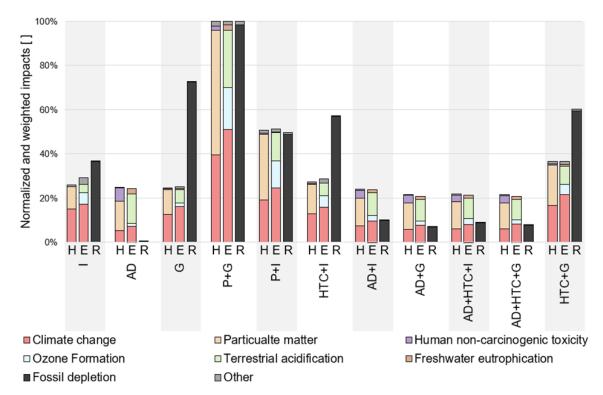


Figure 13: Normalized and weighted results for the considered treatment paths for food waste according to ReCiPe Endpoint (H). H=Damage to human health, E=Damage to ecosystem diversity, R=Damage to resource availability.

Also, for food waste, an in-depth analysis was required to validate the weighted results. A comparative analysis based on the most important impact categories was conducted and is presented in Figure 14. A hotspot analysis is presented alongside to better match the environmental burdens with the causal processes (Figure 15).

#### a. Climate change and fossil depletion

The initial waste collection and transport to the treatment plant, resulted in a base load (combustion of diesel) in all systems. Similarly, the treatment of impurities (modelled as incineration of residual waste) added to this. When waste was pre-stored further greenhouse gases were released. During storage, food waste already underwent biological decomposition and resulted in the release of N<sub>2</sub>O emissions (nitrification/denitrification processes) and methane emissions (anaerobic digestion). Within large scale waste incinerators, it was presumed, that exhaust air from the waste bunker was utilized in the combustion process (primary/secondary combustion air) and emissions resulting from the pre-storage were thus missing in "I" (Figure 15). Further sources for impacts in the respective categories were the prorated consumption of ancillary and capital goods. This justifies the acknowledgment of capital goods and ancillary goods in the modelling phase. Within the considered systems ~10-20% of burdens connected to the GWP and ~25-90% of burdens connected to fossil depletion were caused by ancillary and capital goods.

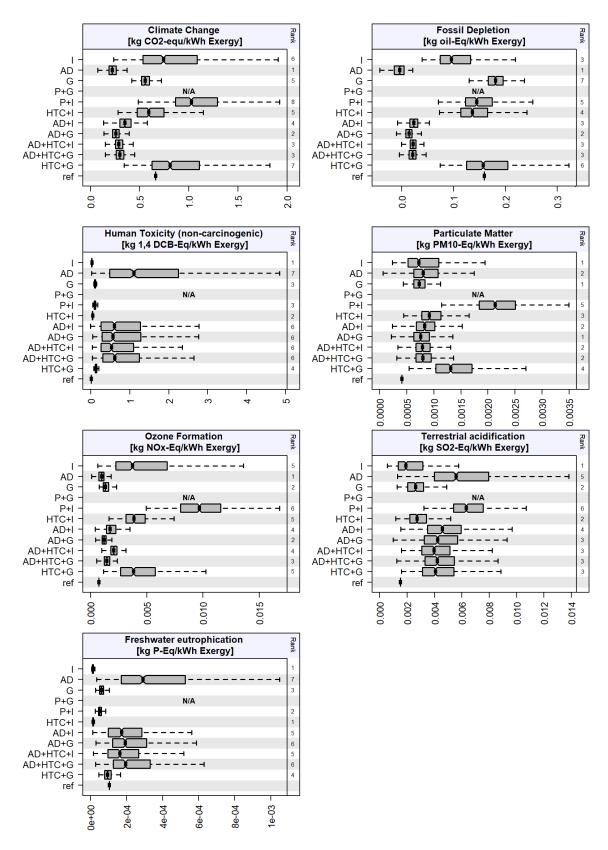


Figure 14: LCIA of the most relevant impact categories for the treatment of food waste. The environmental impacts are presented as notched boxplots, whereby the uncertainty ranges were derived from the Monte-Carlo Simulation. If notches did not overlap, it was implied that the environmental impacts differed significantly. A rank was allocated accordingly (1=lowest impact). The system "ref" accounts for environmental impacts associated with the production of 1 kWh of electricity in the realm of the German electricity mix.

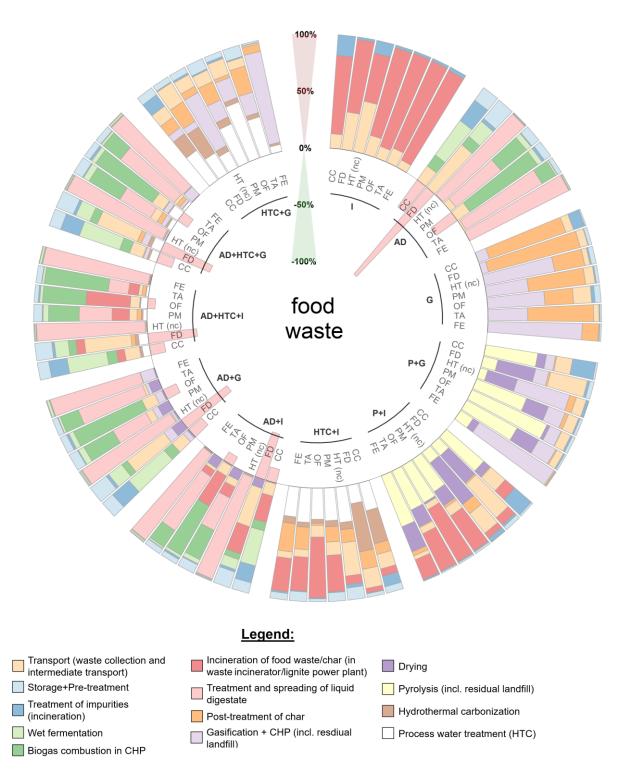


Figure 15: Hotspot analysis of the considered treatment paths for the treatment of food waste. CC=climate change, FD=fossil depletion, HT (nc)=Human toxicity non-carcinogenic, PM=particulate matter, OF=ozone formation, TA=terrestrial acidification, FE=freshwater eutrophication. Results were normalized in a way that no bar exceeded + or -100%.

The system "I" may be classified as autarkic system, but due to the low heating value of the input substrate, the overall system efficiency was reduced, and the respective impacts increased. The GWP and fossil depletion potential was thereby composed of two main sources. Firstly, external heat was needed to pre-heat the flue gas prior to the DeNox stage and secondly, N<sub>2</sub>O-emissions may be formed and released during the waste incineration (as discussed in 8.1.2).

In systems with a high demand for external fuel, the GWP and fossil depletion potential increased strongly due to the high emission and resource consumption factor of natural gas. "G", "P+G", "P+I", "HTC+I" and "HTC+G" required external heat and were not environmentally sustainable with regards to the categories climate change and fossil depletion (Figure 14). In Figure 15 it becomes visible that processes, where support fuel is introduced (e.g., the HTC process or drying), showed a high impact in the hotspot analysis.

Systems that included an anaerobic digestion step were overall characterized by the lowest GWP (Figure 14). The systems ran autarkic and were additionally subject to nutrient recycling, because either all nutrients were recycled (system "AD") or a share of it was returned to agricultural soils via the liquid digestate (systems "AD+I", "AD+G", "AD+HTC+I", "AD+HTC+G"). In turn this step reduced the fossil depletion potential and GWP, as mineral fertilizer (N, P, K) was offset (Figure 15). Detrimental effects associated with anaerobic digestion were caused by CH<sub>4</sub>-slippage during fermentation and biogas combustion (~1% of produced CH<sub>4</sub>).

Additionally, larger amounts of laughing gas ( $N_2O$ ) were released during the spreading of digestate. During nitrification and denitrification processes ~1.25% ( $f_{N2O}$ ) of the applied nitrogen is converted into  $N_2O$ -N. The given value for  $f_{N2O}$  stems mainly from measuring campaigns where mineral fertilizer was applied (PENMAN ET AL. 2000). CUHLS ET AL. (2008) argue that the emission factor of  $f_{N2O}$  is applicable, but emissions are only released from the soluble and mineral fraction of  $N_{total}$ . They therefore suggest that  $N_2O$ -N emissions are calculated according to MFE x  $N_{total}$  x  $f_{N2O}$ , where MFE=mineral fertilizer equivalent. HANSEN ET AL. (2006) modelled the calculation of  $N_2O$ -N emissions from the application of organic residues and calculated the  $N_2O$  emissions according to  $N_{total}$  x  $f_{N2O}$ , where  $f_{N2O}$ =~1.5%. Lastly JUNGKUNST ET AL. (2006), argued that  $N_2O$  emissions from agricultural soils are mainly site-specific and may not necessarily be linked to the nitrogen content of the fertilizer. Given this inconclusiveness, a high uncertainty was introduced to the system.

# b. Human toxicity (non-carcinogenic)

Even though the weighting step suggested that non-carcinogenic human toxicity was of primary importance to the environmental sustainability, an in-depth analysis showed that the respective impacts were negligible. Impacts were primarily a result of the ill-defined methodology which is given in most LCIA methods. A linearity between cause and effect is given, while threshold values are neglected.

Heavy metals which were initially present in the substrate were eventually released to the environment. In the realm of thermochemical treatment systems, they were mainly retained in the flue gas treatment systems. Later they may be released as intermediate emissions from the landfilled ash or slag via leachate flows. The utilized methodology did however neglect long-term effects (the considered time frame was 100 years). When digestate was spread on agricultural areas, all heavy metals present in the substrate were immediately released to the soil (short-term emissions). As seen in Figure 14, the highest potential was given in the system AD (>1 kg 1,4 DCB-Eq/kWh Exergy). The impact arose almost exclusively from the release of zinc which was present in food waste. The emissions factor was hereby ~65 mg Zn/kg DM digestate. TÓTH ET AL. (2016) suggest that ecological/health risk are not to be expected from concentrations <250 mg Zn/kg DM soil and accordingly it appears little plausible that toxicity is provoked by spreading digestate, which was derived from food waste.

#### c. Particulate matter formation

The product systems showed similar results per kg of treated material throughout this category and thus a high correlation to the conversion efficiency of substrate into exergy. Versatile sources of particulate matter were present in the considered treatment systems, most of which were emitted as secondary particulate matter in the form of NH<sub>3</sub>, SO<sub>2</sub> and NO<sub>x</sub>.

NH<sub>3</sub> emissions derived from the storage, drying and spreading of food waste or its digestate. While indoor emissions of NH<sub>3</sub> may be abated by biofilters and/or acidic scrubbers, no mechanisms controlled the release of NH<sub>3</sub> from spreading digestate on agricultural areas. 7.5% of N<sub>total</sub> from the digestate were released as NH3 and therefore, the largest NH<sub>3</sub> emissions were found in the system "AD" (Figure 14).

A thermochemical treatment of food waste volatilized the sulfur in the substrate, which was finally oxidized to  $SO_2$  (and to a much smaller extent to  $SO_3$ ). Flue gas treatment systems removed >>90% of the  $SO_x$  by means of dry sorption and/or wet scrubbers to comply with the limit values of 50 mg/m³ of flue gas (17. BImSchV). Within biogas plants sulfur is initially volatilized as  $H_2S$  and may be found in the raw biogas. During the combustion of biogas,  $H_2S$  would

react to SO<sub>2</sub> and H<sub>2</sub>O. To meet the current limit values of 310 mg SO<sub>x</sub>/m<sup>3</sup> flue gas (44. Blm-SchV, existing gas engines), the biogas was desulfurized prior to combustion (in this model by adding iron-(II)-chloride to the reactor).

NO<sub>x</sub> emissions resulted from the combustion processes of the substrate, biogas or support fuel (Figure 15). Despite a continuous tightening of the legislative framework, NO<sub>x</sub> emissions were the main driver of particulate matter in this framework. Allowable NOx emissions are currently 150 mg/m³ and 200 mg/m³ for the combustion in waste treatment facilities with a combustion performance of >50 MW and <50 MW, respectively. For the combustion of biogas, the limit values are currently 500 mg NO<sub>x</sub>/m³ (existing gas engines).

#### d. Ozone formation and terrestrial acidification

As already discussed in 8.1.2 both ozone formation and terrestrial acidification are closely linked to the category particulate matter formation. A higher characterization factor for NH<sub>3</sub> is given in the category terrestrial acidification in comparison to the category particulate matter. Systems that included a spreading of digestate (where larger amounts of NH<sub>3</sub> were released) thus showed an increased acidification potential in comparison to the quantified potentials in the category particulate matter formation (Figure 14, Figure 15). Ozone formation was for >99% provoked by NO<sub>x</sub> emissions. The amount of emitted NO<sub>x</sub> is similar throughout all systems and thus highly correlated with the amount of treated material (Figure 11).

## e. Freshwater Eutrophication

Eutrophication can take effect during the spreading of digestate. Both the return of P to the nutrient cycle (freshwater eutrophication) and the runoff of nitrate to groundwater (marine eutrophication) hold the potential to cause eutrophication. In the course of weighting, a much higher impact was allocated to freshwater eutrophication in comparison to marine eutrophication (Figure 13). Whether eutrophication becomes relevant in this context, cannot be represented by eLCA. It is dependent on the regional circumstances (nutrient supply, land usage, soil type etc.) and simply put, takes effect, if the amounts of nutrient availability and the plant uptake are imbalanced.

# 8.2.3 Economic assessment

Figure 16 shows the economic assessment of the considered treatment options as violin plots. For all considered treatment systems, the mean total costs, separated for treatment costs and waste collection are given. As the systems "P+G" and "P+I" showed disproportionately high LCOE, they were not presented in the chart. They were not competitive in this framework, due to the exceptionally low exergy output.

Like the environmental balance, the treatment paths, which contained an anaerobic digestion step were characterized by the overall lowest LCOE. Herein, anaerobic digestion followed by spreading ("AD") represented the most profitable option at average total costs of 49 ct/kWh<sub>Ex</sub>. When the digestate was utilized for energy recovery in the realm of a cascaded waste treatment ("AD+I", "AD+G", "AD+HTC+I", AD+HTC+G"), the treatment costs increased on average by 35-60%. The total costs (treatment costs + costs for waste collection) increased by ~15%. This was a result of additional economic burdens for further treatment plants, which could not be compensated by an additional energy output (Figure 14).

Direct thermochemical treatment according to "I" or "G" was found to be less favorable. Both gasification and incineration plants were characterized by higher investment and O+M costs than biogas plants and additionally the specific energy output from both systems was reduced in comparison to "AD". In "I" source segregation was not required within this system as food waste was collected as part of the residual waste. This reduced the economic burdens of waste collection, but the treatment path was still not competitive. The total costs were on average 86 ct/kWh<sub>Ex</sub>.

The system "HTC+I" was also economically inferior to "AD". The overall exergy output was not increased, and the HTC plant held higher lifetime investment and O+M costs than "AD". "HTC+G" showed an even lower exergy exploitation rate than "HTC+I". Lastly, the gasification step was costlier than the co-combustion of HTC-char in a lignite power plant, despite additional transportation costs.

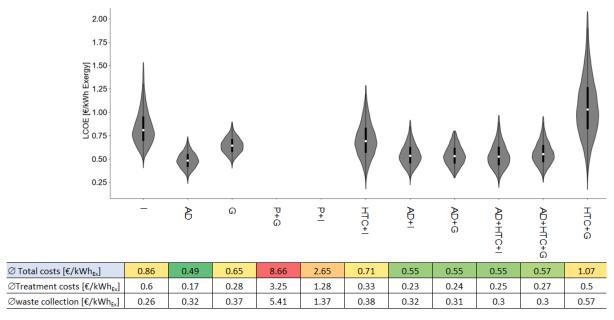


Figure 16: LCOE for different treatment options of food waste. Violin plots represent the distribution curve. Outliers were excluded (Z-score<3). Total costs are given separately for treatment costs and waste collection costs.

#### 8.2.4 Conclusive recommendations

In the light of both the environmental and economic assessment, the treatment path "AD" appeared to be the preferential treatment option for food waste. Also, treatment options that contained one or multiple thermochemical post-treatment steps of the digestate were characterized by a preferential set-up ("AD+I", "AD+G", "AD+HTC+I", "AD+HTC+G"). In comparison to "AD", the cascaded treatment options showed overall similar environmental burdens, but resulted in an increased impact in the category "fossil depletion". Hereby, longer process chains implied a higher demand in ancillary and capital goods. Secondly, the fossil depletion potential could be drastically reduced in "AD" by credits for nutrient recovery from the spreading of digestate. Similar credits could not be achieved in the cascaded treatment systems, as only a share of the digestate was spread on agricultural areas.

The spreading of digestate may however sometimes state an exclusion criterion for "AD", when regionally saturated or oversaturated fertilization conditions on agricultural areas are given. An additional spreading of nitrogen and phosphorous via the digestate may then lead to overall negative effects for ecosystems. Examples according to HAMACHER ET AL. (2021) include:

- terrestrial and aquatic eutrophication and thereby a loss in biodiversity.
- nitrate runoff, as it cannot be absorbed by plant species anymore; this may lead to a leaching of alkali and soil acidification.
- increased nitrate amounts may infiltrate the groundwater. If a threshold value is exceeded, adverse health effects may be the consequence.

If the regional conditions would exclude the application of digestate, a thermochemical post-treatment step is recommended. Against the background of the technological feasibility, the system "AD+I" appeared as the preferential alternative. A thermochemical conversion that includes gasification and/or HTC is equally applicable, but may state a more complex setup, while the environmental and economic burdens are not reduced.

The remaining treatment paths were found to be little sustainable. The treatment paths "P+G", "P+I" and "I" represented options with a disproportionately low energy output. This in turn was the decisive factor that deteriorated the environmental and economic balance. Wet carbonization according to "HTC+I" and "HTC+G" stated a more suitable approach, as the overall energy output was increased. At the same time, the need for a support fuel was required and led to a high fossil depletion potential and GWP. Equally, the direct gasification ("G") of food waste required higher demands of external energy (for pre-drying the substrate) and is therefore not recommended.

#### 8.3 OFMSW

As the treatment paths and substrate qualities are similar for OFMSW and food waste, the focus in this section lies upon the comparison of these two waste streams. This also aids in answering the question, whether the current system for source segregation needs to be extended for food waste.

#### 8.3.1 Exergetic performance

In Figure 17 the "exergy:support fuel" ratio is stated for the considered treatment paths of OF-MSW. When comparing the specific exergy outputs for OFMSW and food waste, the chemically bound energy in the substrates served as initial indicator. The LHV of food waste and OFMSW were 3.8 and 4.3 MJ/kg respectively.

*Incineration:* The direct incineration ("I") of OFMSW resulted in a slightly higher specific exergy output in comparison to food waste (0.16 kWh<sub>Ex</sub>/kg FM<sub>OFMSW</sub> vs. 0.15 kWh<sub>Ex</sub>/kg FM<sub>food waste</sub>), due to the higher LHV of OFMSW.

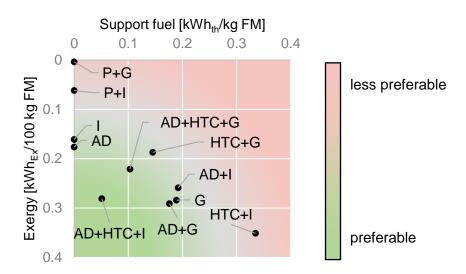


Figure 17: Exergy:support fuel ratio for the considered treatment paths for OFMSW

Anaerobic Digestion: For "AD" the specific exergy output for OFMSW (<0.2 kWh<sub>Ex</sub>/kg FM) was lower in comparison to food waste (>0.3 kWh/kg FM). OFMSW contains a higher share of complex biopolymers (some of which are removed beforehand as structural material), while this is not the case for food waste. The biodegradability of OFMSW during anaerobic digestion was therefore reduced in comparison to food waste. For OFMSW ~45% of the LHV were converted into biogas, while this value was >60% for food waste. Consequently, less high-temperature heat was provided during the biogas combustion. By implication, this reduced the amount of available heat for drying and/or HTC of the digestate, if a thermochemical post-treatment

step was aimed for. In comparison to food waste, the systems "AD+HTC+I", "AD+HTC+G", "AD+I" and "AD+G" were not autarkic anymore and 0.05-0.2 kWh<sub>th</sub> of support fuel per kg FM were required. At the same time, a secondary thermal post-treatment step appeared more meaningful for OFMSW in comparison to food waste, as more chemically bound exergy was available for energy generation. The overall exergy exploitation could be increased by ~50% if the digestate was energetically utilized.

Gasification: the specific exergy output in "G" was similar for both substrates (~0.3 kWh<sub>Ex</sub>/kg FM). Slightly higher cold gas efficiencies were modelled for food waste and thus the slightly lower LHV of food waste in comparison to OFMSW could be compensated. As OFMSW held a lower moisture content in comparison to food waste, the required support fuel for pre-drying was reduced to ~0.2 kWh<sub>th</sub>/kg FM for OFMSW (food waste=~0.4 kWh<sub>th</sub>/kg FM).

HTC: The HTC process showed an overall higher performance for OFMSW in comparison to food waste. Due to a higher share of structural material (e.g. woody biomass), a lower amount of chemically bound water and a lower share of proteins, the product yield of hydrochar from OFMSW was higher (0.64 kg char<sub>DM</sub>/kg biomass<sub>DM</sub>) than that of food waste (0.52 kg char<sub>DM</sub>/kg biomass<sub>DM</sub>). Therefore, the specific exergy output was increased for the systems "HTC+I" and "HTC+G" for the substrate OFMSW in comparison to food waste. When HTC was coupled with anaerobic digestion, support fuel could be pared down, as high temperature heat could be provided from the combustion of biogas. Following this, "AD+HTC+I" emerged to be one of the most desirable options.

*Pyrolysis:* The dry carbonization according to pyrolysis was characterized by the lowest specific exergy outputs. Almost the entire energy that resulted from the combustion of the volatile pyrolysis products (gas+oil) was consumed in the drying process. The char yields were comparably low at <0.4 kg char<sub>dry</sub>/kg OFMSW<sub>dry</sub> and thus the exergy yield from the post-treatment of the char was reduced.

## 8.3.2 Environmental assessment

Like the evaluation of food waste, the same impact categories were of increased relevance for the treatment paths of OFMSW. Those were (i) climate change, (ii) fossil depletion, (iii) human toxicity, (iv) particulate matter, (v) ozone formation, (vi) terrestrial acidification and (vii) freshwater eutrophication. The weighted results are given in Figure 18.

For the systems "P+I" and "P+G" disproportionately high environmental burdens were quantified, due to the limited exergy output. Both "P+I" and "P+G" were therefore excluded from the environmental balance. For the remaining systems, a preferential tendency is at first not given

for either treatment path. Still, some treatment paths appeared little promising. The system "G" was pareto-efficient for all three endpoint categories against the systems "HTC+G", "HTC+I" and "AD+I". On the other hand, "I" appeared to outcompete "G", "AD+G", "AD+HTC+I" and "AD+HTC+G", mainly due to a lower "Damage to resource availability". In conclusion, the systems "I" and "AD+comp" emerged as the most environmental solutions. However, weighting is subjective and the differences between the systems, are little pronounced. Additionally, "AD+comp" showed the highest damage to human health, which may state an exclusion criterion for this path. To disentangle the prevailing ambiguity, the treatment paths were considered in more detail in a comparative and hotspot analysis.

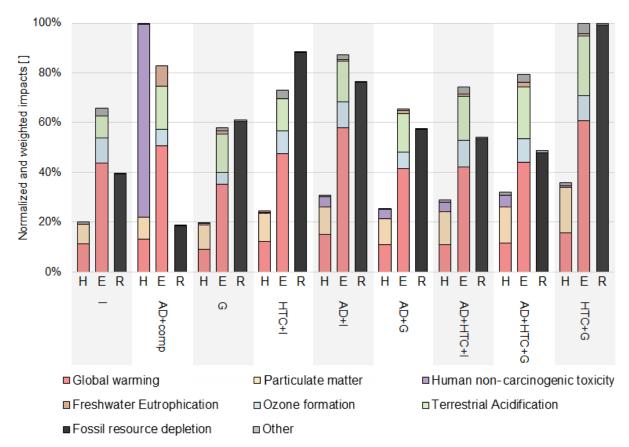


Figure 18: Normalized and weighted results for the considered treatment paths for OFMSW according to ReCiPe Endpoint (H). H=Damage to human health, E=Damage to ecosystem diversity, R=Damage to resource availability.

In Figure 19 the LCIA of the main impact categories is presented and in Figure 20 the hotspot analysis is given. With respect to the evaluation the differences between the treatment of OF-MSW and food waste are here mainly pointed out, rather than discussing effect mechanisms as described in 8.2 again.

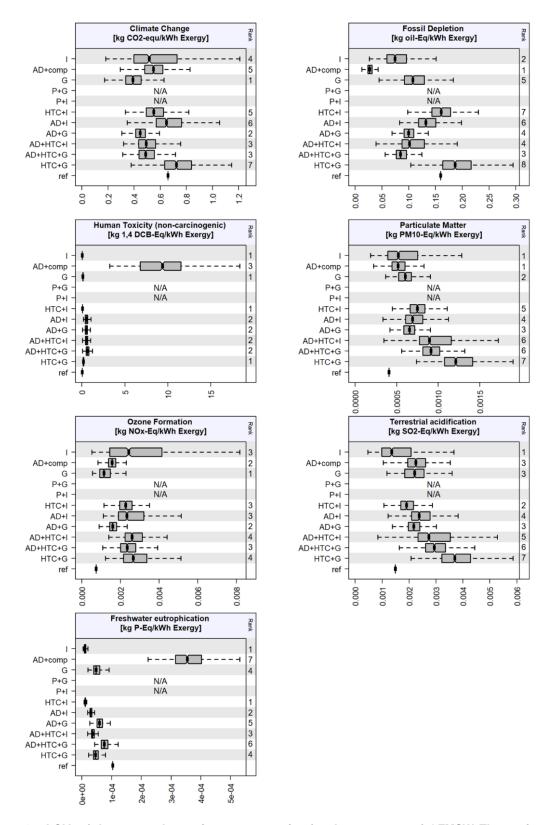


Figure 19: LCIA of the most relevant impact categories for the treatment of OFMSW The environmental impacts are presented as notched boxplots, whereby the uncertainty ranges were derived from the Monte-Carlo Simulation. If notches did not overlap, it was implied that the environmental impacts differ significantly. A rank was allocated accordingly (1=lowest impact). The system "ref" accounts for environmental impacts associated with the production of 1 kWh of electricity in the realm of the German electricity mix.

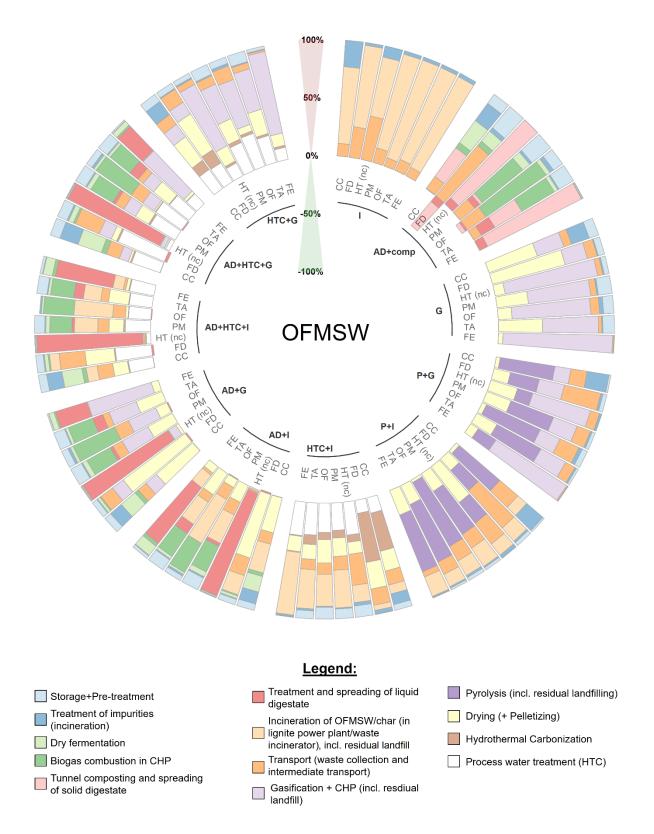


Figure 20: Hotspot analysis of the considered treatment paths for the treatment of OFMSW. CC=climate change, FD=fossil depletion, HT (nc)=Human toxicity non-carcinogenic, PM=particulate matter, OF=ozone formation, TA=terrestrial acidification, FE=freshwater eutrophication. Results were normalized in a way that no bar exceeded + or -100%.

### a. Climate change and fossil depletion

In accordance with the evaluation of the substrate food waste, a high share of fossil depletion was provoked by the combustion of fossil fuels for (i) drying, (ii) the HTC process and (iii) to a smaller extend for the pre-heating of the flue gas prior to the DeNox-stage in incinerators (Figure 20). In this context, two main differences prevailed between OFMSW and food waste. Firstly, the water content of raw OFMSW was lower in comparison to food waste. This reduced the amount of support fuel for pre-drying substrates (e.g. required for the system "G"). Secondly, as discussed in 8.3.1, additional support fuel was required for cascaded treatment systems that contained anaerobic digestion ("AD+I", "AD+G", "AD+HTC+I", "AD+HTC+G").

Further differences resulted from the decreased amount of nitrogen in OFMSW. Thereby the credits for spreading digestate were reduced, as less soluble and mineral nitrogen was transferred to agricultural areas. Secondly, the reduced share of nitrogen led to fewer emissions of N<sub>2</sub>O, which were emitted during the storage of substrate, the spreading of digestate or the combustion of OFMSW or its derivatives.

Finally, when the mono-treatment according to anaerobic digestion is compared for OFSMW and food waste, fundamental discrepancies arose. The respective treatment paths were represented by "AD+comp" (OFMSW) and "AD" (food waste). For OFMSW, the digestate underwent a solid-liquid separation step and the solid digestate was composted. For food waste, the digestate was directly spread without further treatment. The composting step led to additional greenhouse gas emissions in the form of  $CH_4$  (anaerobic conditions in the rotting units due to oxygen deficiencies) and  $N_2O$  (leakage currents of the nitrification and denitrification processes). When the exhaust air passes the biofilter, the amount of  $N_2O$  emissions may increase further, as a share of  $NH_3$  (major emission from composting plants) is converted into  $N_2O$  (CUHLS 2012). Therefore, the installation of an upstream acidic scrubber is highly recommended. The emissions could be further reduced, if a share of the exhaust air was used as combustion air for the biogas, but this was not implemented. As a second implication of the composting >90% of the nitrogen is initially organically bound. This decreased the credits for mineral fertilizer replacement according to the MFE but may also decrease the  $N_2O$  emissions, as discussed in 8.2.2b.

## b. Human toxicity

The share of heavy metals was increased in OFMSW in comparison to food waste. The disproportionately large impact, which was given for "AD+comp" (Figure 19) must be critically viewed. The resulting heavy metal inputs to the soil undercut the limit values for heavy metals as given in the BioAbfV. Additionally, the application of digestate from OFMSW is currently

restricted to 20 t of DM/ha within three years (BioAbfV), which would curb an accumulation of heavy metals resulting from OFMSW application. On this account, KNAPPE ET AL. (2012) analyzed the toxicity resulting from composted OFMSW. They stated that an exceedance of the precautionary values stated in the Federal Directive on Soil Purity (BBodSchV) is not to be expected and toxic effects from spreading of OFMSW derivatives will not take effect. This is equally true for long-term spreading of OFMSW derivatives.

### c. Particulate Matter, Ozone Formation and Acidification

In comparison to the spreading of digested food waste ("AD"), the amount of uncontrolled NH<sub>3</sub> emissions was reduced when OFMSW was spread on agricultural areas ("AD+comp"). The digestate from OFMSW contained a lower nitrogen content and was mainly spread as compost. Compost holds a much lower ammonia content (1% of N<sub>total</sub>) in comparison to liquid digestate (50% of N<sub>total</sub>) (HANSEN ET AL. 2006).

Initially, the lower share of nitrogen in OFMSW also led to lower NO<sub>x</sub> emissions during thermochemical treatment steps. During incineration of OFMSW ("I") or its digestate ("AD+I") the mass-specific NO<sub>x</sub> emissions were reduced by ~30% in comparison to the respective pathways for food waste. This linear approach may be applied for large-scale incinerators, where the share of OFMSW or food waste would only account for a small fraction of the total combusted waste. It was assumed that higher NO<sub>x</sub> emissions resulting from the combustion of OFMSW or food waste could be compensated by other waste fractions, so that the regulations of the 17. BlmSchV could be complied with. For gasification and pyrolysis this mindset was not applicable. There, only one waste fraction (i.e. OFMSW and food waste) was treated and higher NO<sub>x</sub> emissions resulting from OFMSW or food waste could not be compensated by other waste fractions. For these setups, the NO<sub>x</sub> emissions were strictly adjusted to the statutory regulations and similar emissions were incorporated for OFMSW and food waste.

### d. Freshwater eutrophication

Similar to food waste, a large eutrophication potential could be determined during the spreading of digestate. Also, for OFMSW, freshwater eutrophication is provoked by the return of P to the nutrient cycle. This impact may be disrespected, if it is assumed that digestate is spread in a needs-oriented manner.

### 8.3.3 Economic assessment

Figure 21 states the results of the economic assessment. The overall lowest economic burdens were found for "AD+G" at ~60 ct/kWh exergy. The conversion efficiency of OFMSW into exergy during "AD+G" was higher in comparison to "AD+comp". This decreased both the treatment

and waste collection costs. As anaerobic digestion was characterized by lower lifetime expenses than gasification, the initial anaerobic digestion step was economically worthwhile. "AD+G" showed LCOE that were ~20% lower than those of "G", while both systems produced similar amounts of exergy. Additionally, a high share of heat, which was required for the drying of the digestate could be provided by on-site CHP plants and costs for external fuel were reduced.

"HTC+I" represented a system with an equally high economic performance. In comparison to "AD+G", the energy conversion was increased. The lifetime expenses for the infrastructure in both systems was comparable, but since a secondary transportation from the first treatment plant (HTC-plant) to the lignite power plant was required, the LCOE of "HTC+I" were not reduced in comparison to "AD+G".

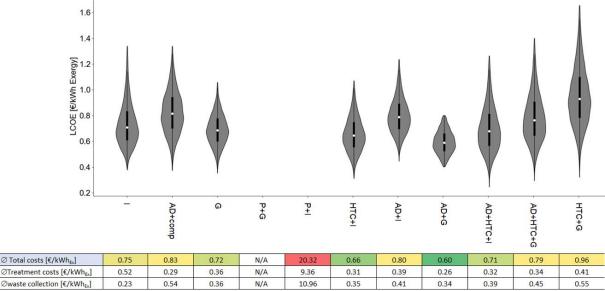


Figure 21: LCOE for different treatment options of OFMSW. Violin plots represent the distribution curve. Outliers were excluded (Z-score<3). Total costs are given separately for treatment costs and waste collection costs.

The remaining systems were less favorable, but - apart from "P+I" - not characterized by disproportionally high LCOE.

- Direct incineration ("I") showed high treatment costs (75ct/kWh), due to the low energy conversion efficiency of the system. However, as source segregation was not required for this path, the waste collection costs could be decreased.
- For "AD+comp" the opposite effect was given. The treatment costs were comparably low (29 ct/kWh), although a composting plant was additionally needed. However, as the energy content of the digestate was not utilized, an overall low energetic exploitation potential was the consequence and therefore the specific waste collection costs increased to 54 ct/kWh.

- Combustion of the digestate according to "AD+I" would increase the conversion efficiency into energy, but for the incineration of the digestate additional support fuel for drying (to allow for an autothermal combustion) and transportation to a centralized incineration plant was required.
- The gasification of HTC char ("HTC+G") achieved a lower exergy exploitation than incineration ("HTC+I") and making the system less competitive.
- "AD+HTC+I" featured three conversion steps and the economic expenses were increased by ~10% in comparison to "HTC+I". Additionally, the specific exergy production was reduced in comparison to "HTC+I".
- In "AD+HTC+G", the higher specific exergy production in comparison to "HTC+G" could compensate the higher economic burdens which were required for three treatment systems. The LCOE was decreased by ~20% in comparison to "HTC+G".

### 8.3.4 Conclusive recommendations

The treatment paths "I", "AD+comp" showed the highest environmental performance. Given the large uncertainties also "G", "AD+HTC+I" and "AD+HTC+G" appeared meaningful. Since the treatment path "AD+HTC+I" showed lower LCOE, but otherwise similar environmental impacts, it was found to be preferable over "AD+HTC+G". Further selection of the treatment paths will and must not be performed. From this point onward the generalist approach, which was used in this work, reaches its limits. The question, how OFMSW should be treated, must be answered according to regional circumstances:

- (1) Is a need for fertilizer from OFMSW given in the region? If this is the case, the credits for fertilizing, which were included in the eLCA and LCOE are justified and "AD+comp" is always recommended. From an eLCA point it is argued that the system "AD+comp" is highly attractive due to its disproportionately low fossil depletion potential. From a circular economy point it may be argued that this treatment is beneficial as it combines recovery with recycling and at the same time the key aspect of the conservation of resources is maintained.
- (2) What is the technological feasibility of the treatment systems at the time of implementation? Especially the energy-related values and mass distributions were based on models and lab-scaled experiments for gasification and HTC. For large-scale plants, the performance may be impaired. In case that a thermochemical post-treatment is required, it is questionable, why gasification ("G") should be chosen over incineration ("I"), as the environmental impacts are not reduced.

(3) Where is the point of the waste collection located? Two aspects fall in this category. Firstly, if spatial proximity to a lignite power plant is given, then "AD+HTC+I" becomes an interesting alternative. Transportation efforts are minimized and both the environmental and economic balance are enhanced. The second aspect is associated with the waste composition. While the composition of OFMSW may be shifted towards a higher share of food waste in urban areas, higher shares of green waste are generally found in rural areas (KNAPPE ET AL. 2015). The latter would increase the share of lignocellulosic material in OFMSW, which is conducive for a treatment by means of incineration.

In connection with the last point, it should also be discussed, whether the current form of source segregation must be reconsidered. While OFMSW is currently collected separately, this is not the case for food waste. The preferential treatment path was initially identical for both substrates: a monotreatment according to anaerobic digestion. Thus, this would not justify a separate collection of food waste from OFMSW. If the return of nutrients to nearby agricultural areas would however negatively affect the quality of ecosystems (e.g., through nitrate or phosphate runoff) or if the limit values with regards to toxic elements as stated in the Fertilizer Ordinance are exceeded, the system "AD+HTC+I" represents an approach that results in an overall good balance for both substrates. Alternatively, if only conventional treatment systems are available (e.g., due to the existing infrastructure), it is recommended that green waste is disposed of in residual waste bins, while only food and kitchen waste are source segregated. It is indicated (although not quantified in this study) that the removal of food waste from OF-MSW would enhance the performance of the incineration process of OFMSW.

## 8.4 Sewage sludge

## 8.4.1 Exergetic performance

In Figure 22 the "exergy:support fuel" ratio for the considered treatment paths for sewage sludge is stated. The reference unit was set to 100 kg of raw sewage sludge at a DM-content=2 wt.-% (referred to as FM in the subsequent analyses).

Anaerobic Digestion + spreading: In "AD" exergy could only be harvested during the sewage gas combustion, while the chemically bound energy in the digestate was not utilized for energy recovery. At the same time energy intensive post-treatment steps could be omitted. The specific exergy output was ~1.2 kWh<sub>Ex</sub>/100 kg FM. A smaller share of support fuel was inventoried (~0.15 kWh<sub>th</sub>/kg FM), to maintain the hygienization process.

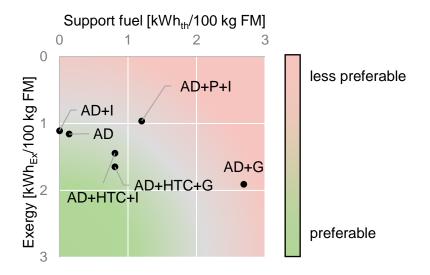


Figure 22:Exergy:support fuel ratio for the considered treatment paths for food waste.

Incineration: In "AD+I" the heat demand for drying could be satisfied by the incineration process and the system ran autarkic. This in turn reduced the net electricity output during incineration to a negligible share. Therefore, the exergy output in "AD+I" was similar to that of "AD".

Gasification: "AD+G" showed the highest specific exergy output (~2 kWh<sub>Ex</sub>/100 kg FM.) as it converted sewage sludge at a high efficiency into heat and electricity. The high efficiency of the gasification process was however "purchased" at the expense of support fuel. The required heat for pre-drying the sewage sludge prior to the gasification could only partly be satisfied by the gasifier. The share of support fuel amounted to 2.7 kWh<sub>th</sub>/ 100 kg FM.

HTC: The implementation of the HTC process as an intermediate process step could reduce the heat demand in comparison to direct drying. To dry sludge from DM=25% (dewatered sludge) to DM=40% (according to "AD+I"), a heat demand of ~2 kWh<sub>th</sub>/100 kg FM was required. For the HTC process (according to "HTC+I" and "HTC+G") the heat demand was only ~0.8 kWh<sub>th</sub>/100 kg FM. After the HTC, the product could be dewatered to a DM-content of 60%. At the same time, it must however be considered that ~30% of the chemically bound energy of the sewage sludge was transferred to the process water/gas and was not available for energy exploitation anymore. The latter aspect reduced the exergy output of "AD+HTC+G", in comparison to "AD+G" from 1.9 to 1.65 kWh<sub>Ex</sub>/100 kg FM.

When comparing "AD+I" and "AD+HTC+I" it was found that the specific exergy output increased from 1.1 to 1.4 kWh<sub>Ex</sub>/100 kg FM. In "AD+HTC+I", the HTC process was performed decentralized and heat was not extracted from the incineration process anymore but provided by support fuel (0.8 kWh<sub>Ex</sub>/100 kg FM). When the HTC-char was incinerated more heat and electricity could be produced, as high temperature heat from the incineration process was not

diverted to a drying unit anymore. Therefore, the higher efficiency of this product system was again purchased at the expense of support fuel.

*Pyrolysis:* The implementation of a pyrolysis step ("AD+P+I") resulted in the overall lowest specific exergy output (<1 kWh<sub>Ex</sub>/100 kg FM). Additionally, external heat in the order of 1.2 kWh<sub>Ex</sub>/100 kg FM was required for drying purposes. The cost-benefit ratio between the energy demand (for drying and pyrolysis) in comparison to the energy output was unfavorable.

### 8.4.2 Environmental assessment

For sewage sludge, two fU were assessed. In alignment with the goal and scope of this work, the production and export of 1 kWh of exergy was referenced as fU and secondly, the fU was set to 100 kg of raw sewage sludge. Initially, the most relevant impact categories were determined for both fU. According to the weighting step, (i) climate change, (ii) fossil depletion, (iii) non-carcinogenic human toxicity, (iv) particulate matter, (v) ozone formation, (vi) terrestrial acidification and (vii) freshwater eutrophication showed the highest relevancy in the evaluations. An individual analysis of the remaining impact categories was not found to lead to further insights, either because the analyses would be redundant (e.g., human toxicity and ecotoxicity are both driven by heavy metals which are released from the systems to the environment) or because they stemmed explicitly from the background system.

In Figure 23, the weighted results are presented for both fU. Given that the balancing approach differed with regards to the allocation procedure for both fU, the quantities within the LCIA were bound to change accordingly. It was found that the patterns within the different impact categories were similar for both fU. This suggests a high robustness of the model and indicates that either approach is valid for the assessment of sewage sludge. One exception was the endpoint category "damage to resource availability". For the mass-based fU (fU=100 kg FM of sewage sludge), exported heat was given a high credit. Therefore, the systems "AD+P+I" and "AD+HTC+I", both of which exported heat to a district heating system, achieved higher benefits in comparison to the assessment, where heat was credited according to its exergy factor.

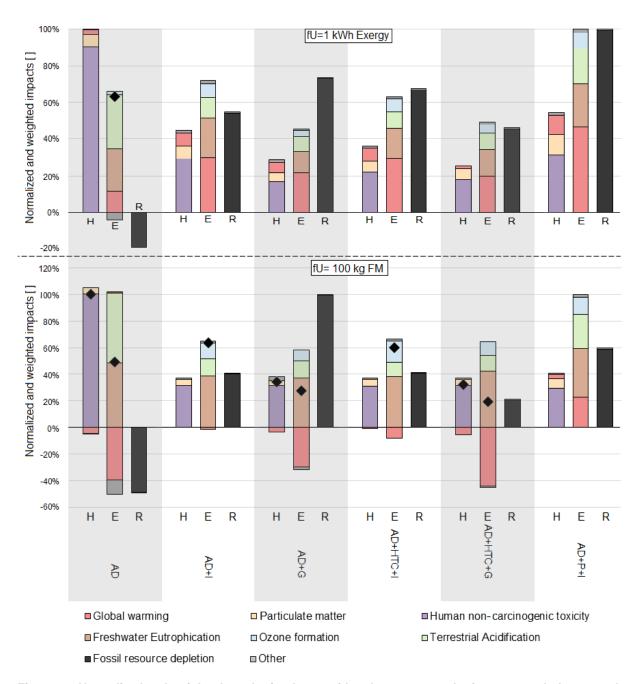


Figure 23: Normalized and weighted results for the considered treatment paths for sewage sludge according to ReCiPe Endpoint (H). H=Damage to human health, E=Damage to ecosystem diversity, R=Damage to resource availability. The upper part represents results for a mass based fU, i.e., input of 100 kg fresh sewage sludge, DM-content=2% and lower part shows results for an exergy based fU, i.e., production and export of 1 kWh of exergy. Diamonds indicate the total burdens.

Based on the weighted results, a macro-analysis of the environmental burdens could be conducted.

- For systems that contained a thermochemical post-treatment step, the overall lowest burdens could be allocated to the system "AD+HTC+G" with regards to both fU.
- "AD+G" showed similar impacts as "AD+HTC+G" but was characterized by an increased "Damage to resource availability".

- In comparison to gasification, incineration appeared to be a less suitable post-treatment option. Both "AD+I" and "AD+HTC+I" were inferior in comparison to the systems "AD+G" and "AD+HTC+G".
- The pyrolysis of sludge was inferior in comparison to HTC. A direct comparison between the systems "AD+P+I" and "AD+HTC+I" attested higher environmental burdens to the pyrolysis process in comparison to the HTC process for most environmental impacts.
- Direct spreading of digested sewage sludge (system "AD") showed the lowest potentials for global impacts such as fossil depletion and climate change. At the same time, the direct spreading of sewage sludge held the highest potential to induce local impacts such as human toxicity, eutrophication and terrestrial acidification.

Whether the weighting step gave a realistic reflection of the environmental impacts, was deduced from an individual evaluation of the respective impact categories. In Figure 24 the LCIA of the most relevant midpoint categories is presented. The results are discussed in conjunction with the hotspot analysis, which is given in Figure 25.

### a. Climate change and fossil depletion

In alignment with the previous assessments in 8.1-8.3, climate change and fossil depletion were closely interlinked. As all systems recovered P, environmental savings could be achieved by offsetting the respective mineral fertilizer (Figure 25). System which contained a thermochemical post-treatment step and recovered P from the ash could offset mineral rock phosphate at an efficiency of ~65% (P-recovery rate x fertilizer efficiency/(P in sludge)). In "AD" the efficiency of the P-recovery was 100% and additionally, conventional N and K fertilizer could be offset. Therefore, the highest savings from nutrient recovery could be achieved in "AD" (Figure 25).

For the alternate fU (treatment of 100 kg FM), heat and electricity, which were produced and exported, were additionally offset. Credits were -0.16 kg  $oil_{Eq}$  and -0.65 g  $CO_{2,Eq}$ /kWh of electricity<sup>3</sup> and -0.08 kg  $oil_{Eq}$  and -0.20 g  $CO_{2,Eq}$ /kWh of heat. The amount of savings went by the amount of produced and exported heat and electricity (Figure 22).

<sup>&</sup>lt;sup>3</sup> These values were extracted from the ecoinvent database. Thereby the reference year was 2016 and the value of 650 g/kWh<sub>el</sub> does not comply with the specific CO<sub>2,Eq</sub>-factors for the electricity mix of the respective year (2020: ~400 g/kWh<sub>el</sub>).

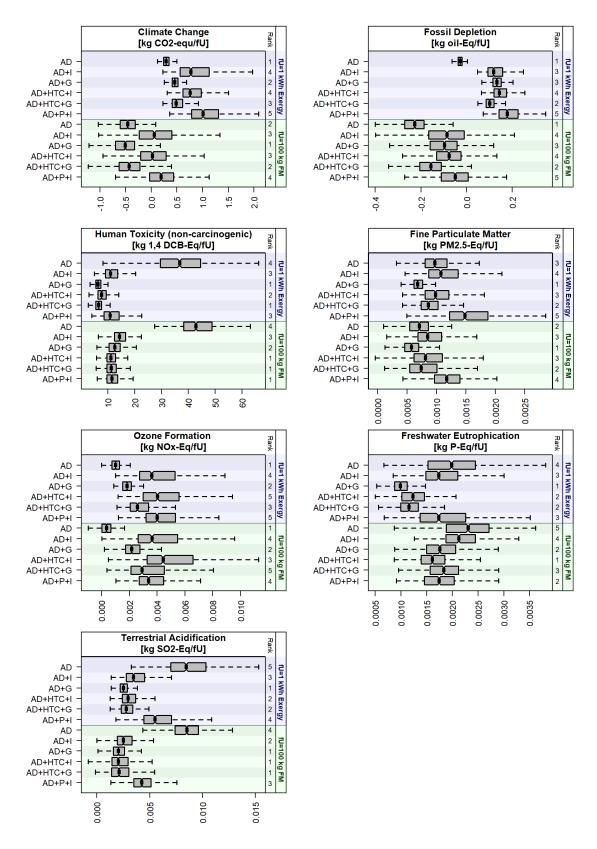


Figure 24: LCIA of the most relevant impact categories for the treatment of sewage sludge. Two fU according to the labels are presented. The environmental impacts are presented as notched boxplots, whereby the uncertainty ranges were derived from the Monte-Carlo Simulation. If notches did not overlap, it was implied that the environmental impacts differ significantly. A rank was allocated accordingly (1=lowest impact). The system "ref" accounts for environmental impacts associated with the production of 1 kWh of electricity in the realm of the German electricity mix.

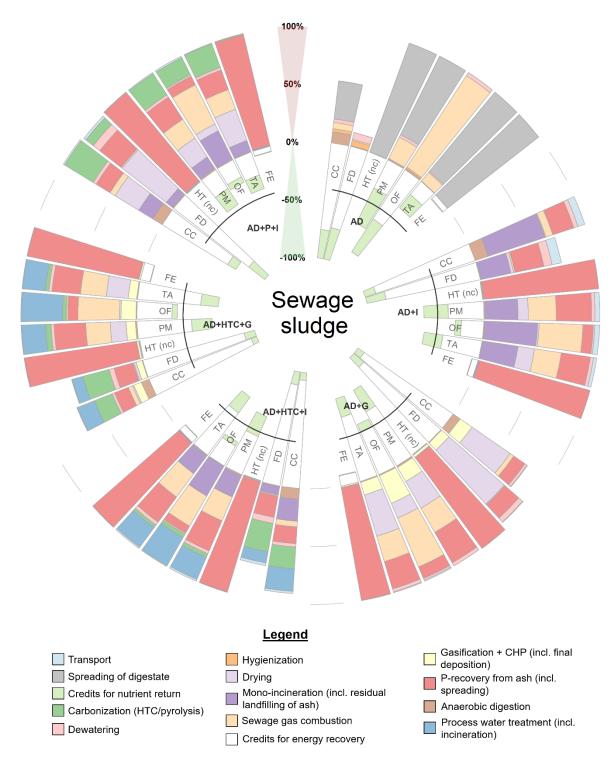


Figure 25: Hotspot analysis of the considered treatment paths for the treatment of sewage sludge. CC=climate change, FD=fossil depletion, HT (nc)=Human toxicity (non-carcinogenic), PM=particulate matter, OF=ozone formation, TA=terrestrial acidification, FE=freshwater eutrophication. Results are normalized in a way that the burdens/credits amount to ±100%. Credits for energy recovery were only applicable, if a fU of "100 kg FM" was considered.

Throughout all systems, thickening, anaerobic digestion, the combustion of the produced sewage gas and dewatering were part of the systems. They led to a base load of ~0.19 kg CO<sub>2,Eq</sub>/100 kg of raw sewage sludge. Main contributors were leakages and slips of CH<sub>4</sub>, during the anaerobic digestion process and the sewage gas combustion. To a smaller extent the consumption of ancillary and capital goods added to these burdens. The fossil depletion potential was little affected by these initial treatment steps and amounted to ~0.02 kg oil<sub>Eq</sub>/100 kg of raw sewage sludge (Figure 25).

In the system "AD" the overall net balance for the category fossil depletion was negative (regardless of the fU), as the return of nutrients could compensate the burdens resulting from the demands of ancillary and capital goods along the process chain (Figure 24 and Figure 25). For the GWP, however a significant burden resulted from the spreading of sewage sludge and more precisely from the emissions of N<sub>2</sub>O (similar to the assessments in 8.2 and 8.3).

The problematic issue of  $N_2O$  emissions is also given for the combustion of sewage sludge and its derivatives (systems "AD+I", "AD+P+I", "AD+HTC+I"). In comparison to grate incinerators, fluidized bed incinerators are known to result in an increased  $N_2O$  emissions (CALVO BUENDIA ET AL. 2019; NEUWAHL ET AL. 2019). STÖCKLEIN ET AL. (2018) evaluated the  $N_2O$  emissions for the combustion of sewage sludge for several fluidized bed incinerators and found that the emissions lie between 470-1480 mg  $N_2O/kg$  DM of sewage sludge. Lower combustion temperatures are applied for fluidized bed incinerators to undercut the ash melting temperature, which in turn increases a potential  $N_2O$  formation. Respectively high emissions with a high uncertainty were introduced in the model, which becomes clearly visible in Figure 24. In this context it may be argued that an initial HTC process may elevate the ash melting point and thus allows for higher combustion temperatures and lower  $N_2O$  emissions. Due to scarcity of data on the factual ash melting point, a combustion at higher temperatures was not implemented. For fluidized bed gasification, the formation of  $N_2O$  is equally enabled, but the post-combustion of the syngas at high temperatures would again not lead to significant  $N_2O$  emissions.

In systems with a thermal post treatment additional burdens stemmed from the consumption of natural gas (as discussed in 8.4.1) and the recovery of P from the ash (Figure 24). The high demand in chemicals and ancillary goods required for the extraction of P entailed burdens that were higher than the savings, which were achieved for the recovery of P. Savings for P-recovery resulted in  $\sim$ -0.04 kg oil<sub>Eq</sub> and  $\sim$ -135 g CO<sub>2,Eq</sub>, while the burdens totaled to  $\sim$ 0.06 kg oil<sub>Eq</sub> and  $\sim$ 300 g CO<sub>2,Eq</sub> per 100 kg of fresh sewage sludge. This becomes visible in the hotspot analysis (Figure 25).

The transportation of sewage sludge to a centralized incineration unit (systems "AD+I", "AD+HTC+I" and "AD+P+I") showed initially comparably low impacts (Figure 25). A distance of 50 km was chosen. The transportation of dewatered sludge to the incinerator resulted hereby in the highest burdens, as it still contained a large share of water (75 wt.-%). This was the case for "AD+I". For larger distances, this treatment path becomes increasingly unattractive, as the burdens increase disproportionately. Vice versa, the transportation effort can be reduced by 70-85% if sewage sludge is carbonized at the site of the WWTP.

The direct comparison between the six product systems showed the lowest impacts for GWP and fossil depletion for the system "AD". "AD+HTC+G" stated the second-best option. In comparison to "AD+G" the implementation of the HTC-step could diminish the external heat demand significantly. For both fU "AD+HTC+I" and "AD+I" showed a slightly lower performance. Firstly, the exergy conversion factor in incinerators was lower than in gasification plants. Secondly negligible N₂O-emissions were balanced for the gasification step. "AD+P+I" was not found to be competitive and wet carbonization according to the HTC-process represented a better alternative.

### b. Human toxicity (non-carcinogenic)

For sewage sludge, the direct spreading of digestate according to the system "AD" resulted in the highest impacts in this category. The LCIA identified zinc and (to a smaller extent) cadmium as main contributors. Also, in this case, eLCA does not provide an appropriate tool to determine toxicity, which is provoked by the spreading of residues on agricultural areas. Sewage sludge represents a highly contaminated waste stream and its application on agricultural soils is therefore regulated by German statutes. Limit values with regards to the concentrations of toxic compounds in the sludge and the maximum spreading quantities (i.e., 5 t of sewage sludge (DM) within three years) restrict the application. In Table 2 the heavy metal concentrations of the referenced sewage sludge are contrasted against the current limit values. It was found that the composition of the sludge used in this study would exceed the limit values for arsenic, cadmium and lead and its spreading would be prohibited. As a second indicator, Table 2 also contains guideline values that would cause either ecological and/or health risks for human beings. These values refer however to the concentration in the soil.

Table 2: Concentrations in sewage sludge in comparison to the limit values, admissible contamination loads and guideline values.

Element	Concentration in di- gested sewage sludge [mg/kg DM] <sup>a</sup>	Limit values in Ger- many [mg/kg DM] <sup>d</sup>	Lower guideline value [mg/kg DM] <sup>e</sup>	Higher guideline value [mg/kg DM] <sup>e</sup>
Arsenic (As)	68	40 <sup>b</sup>	50 (e)	100 (e)
Cadmium (Cd)	6.4	1.5 <sup>b</sup>	10 (e)	20 (e)
Cobalt (Co)	8.0	-	100 (e)	250 (e)
Chromium (Cr)	145	-	200 (e)	300 (e)
Copper (Cu)	296	900 <sup>b</sup>	150 (e)	200 (e)
Mercury (Hg)	3.0	1 <sup>b</sup>	2 (e)	5 (e)
Nickel (Ni)	77	80 <sup>b</sup>	100 (e)	150 (e)
Lead (Pb)	184	150 <sup>b</sup>	200 (t)	750 (e)
Zinc (Zn)	787	4,000°	250 (e)	400 (e)

a) According to values used in this study, b) according to the Fertilizer Ordinance (DüMV), c) according to the Sewage Sludge Ordinance (AbfKlärV), d) Limited to the application of 5 t/ha of sewage sludge in three years e) Lower and higher guideline values according to Tóthet Al. (2016) on heavy metals in soils that would either cause an ecological (e) or health risk for human beings (t)

For an approximate calculation the concentrations in sewage sludge were harmonized with the guideline values for concentrations of heavy metals in the soil. Hereby it was assumed that 5 t DM/ha of sewage sludge were spread within three years. The affected soil depth was set to 20 cm (topsoil) and the density of the soil was  $\rho$ =1.5 t/m³ (dry bulk density). The background concentration of the soil and accumulation effects were at first not taken into consideration. The results are displayed in Figure 26 and may be understood as heavy metal concentrations in the soil after a single application of sewage sludge. The evaluation was performed for the referenced sludge in this study (Sludge 1) and for sludge that showed the maximum admissible concentrations (Sludge 2).

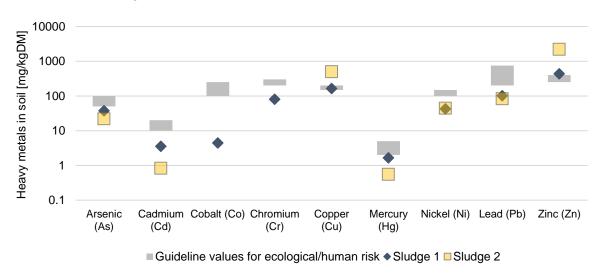


Figure 26: Heavy metal concentrations in the topsoil after a single application of sewage sludge (5 tDM/a within 3 years) contrasted against guideline values (if applicable) that would cause an ecological or health risk.

The rough calculation showed that ecological/health risks may be provoked by both zinc and copper. Enrichment of both heavy metals appears plausible, if sludge is applied multiple times, as (i) zinc and copper are considered little soluble and mobile and (ii) because the removal

rate of heavy metals through soil erosion (Cu=81 g/ha/a, Zn=338 g/ha/a) and harvesting (Cu=34 g/ha/a, Zn=173 g/ha/a; average for typical crop species) falls below the heavy metal entries (BLUME ET AL. 2010). This implies that the current limit values for zinc and copper are too high.

More profound insights could be derived from a risk assessment, but this was beyond the scope of this work. In KRAUS ET AL. (2019) potential toxicity effects from sewage sludge were investigated against the background of toxicity. It was found that the concentrations for zinc and copper would be significantly increased if sewage sludge was used as sole source for fertilizer purposes. However, they concluded that the risk for human beings would "most probably still remain at an acceptable range". In the context of this study, also the toxicity for soil organisms and groundwater was assessed. For soil organisms an increased risk (deriving from zinc and copper) was determined. Regarding groundwater contamination, an elevated risk resulted from copper and to a lesser extent from lead and nickel. Finally, also toxic organic substances were investigated. In this regard KRAUS ET AL. (2019) determined that the biological degradation rates were high enough to rule out toxic effects resulting from pharmaceuticals (negligible effects) and nonpolar pollutants (acceptable risk) for both soil and groundwater. This was equally true for a long-term application of sewage sludge.

For systems that contained a thermal post-treatment step, the transfer of heavy metals to agricultural areas was still relevant. Depending on the recovery strategy, a depletion in heavy metals in the P-product may not be given (e.g. in the approaches AshDec, RecoPhos or LeachPhos) and similar mechanisms as described above are applicable. Overall, the human toxicity potential was found to be lower, when P was recovered from the ash instead of direct spreading of sewage sludge, because not 100% of the initial P was recovered and because some approaches for P-recovery resulted in a depolluted product.

## c. Particulate Matter, Ozone Formation and Acidification

The line of argumentation for particulate matter, ozone formation and acidification is very similar to those given in chapters 8.1-8.3. The initial process steps, which were the same for all product systems (i.e., thickening, anaerobic digestion, sewage gas combustion and dewatering), led to the emissions of 1.5 g of NO<sub>x</sub> and 0.5 g of SO<sub>x</sub>. Most of these emissions could be allocated to the combustion of sewage gas (Figure 25). For systems that contained a thermal post-treatment step, mass-specific NO<sub>x</sub> and SO<sub>2</sub> emissions were in the same range for incineration, gasification and pyrolysis, because similar flue gas systems were given and because all treatment paths were outlined towards the regulations of the 17. BlmSchV. The additional NO<sub>x</sub> emissions ranged between 0.5 and 1.8 g of NO<sub>x</sub> per 100 kg of raw sewage sludge. An

upstream HTC or drying step reduced the NO<sub>x</sub> emissions from this step, as less mass was treated and therefore less flue gas was produced (while the NO<sub>x</sub> emissions per m³ of flue gas stayed constant). At the same time NO<sub>x</sub> and SO<sub>x</sub> emissions were released during the combustion of HTC-process water and the combustion of support fuel. Finally any consumption of capital and ancillary goods was also associated with the release of NO<sub>x</sub> and SO<sub>x</sub> emissions in the background system. NH<sub>3</sub> emissions, which compromised the acidification potential and to a smaller extent the particulate matter formation potential were released during the drying and most importantly during the spreading of sewage sludge.

Within the considered impact categories, "AD+G" could slightly outcompete all other systems, with a thermochemical post-treatment step. While the mass-specific emissions of NO<sub>x</sub> and NH<sub>3</sub> and SO<sub>x</sub> were similar throughout the considered systems, the credits acquired from offsetting exergy were the highest in the system "AD+G". This was the decisive edge in this context. The system "AD" showed the lowest NO<sub>x</sub> emissions, but the highest NH<sub>3</sub> emissions (resulting from the spreading of digestate). As a consequence, the particulate matter formation potential was similar to those of the other treatment paths, the ozone formation potential was reduced and the terrestrial acidification potential was much higher in comparison to other systems (Figure 24).

### d. Freshwater eutrophication

Impacts in the category freshwater eutrophication were almost exclusively a result of the return of P to agricultural areas (Figure 25). The system "AD" showed the highest P recovery rate, i.e., 100% with regards to the share of P in the sludge and thus the highest impact in this category (Figure 24). Since the recovery of P was a core objective for the treatment of sewage sludge, it appears implausible to allocate environmental burdens to the application of P. For systems with a thermal post-treatment step, P was recovered in its pure form. Limitations in place-bound spreading of digestate are practically neutralized, as the transportation effort can be drastically reduced. If P was spread as part of the digested and dewatered sewage sludge, ~150 kg of sewage sludge per 1 kg of P would have to be transported. In other words, the transportation effort can be reduced by the factor 150 if P was spread in its pure form. In compliance with the amended EU fertilizer regulation, the Fertilizing Products regulation ((EU) 2019/1009), from 2022 onwards it will further be possible on to trade organic-based fertilizers within the European Union. This extends the application of P to areas far beyond the location of the production site.

### 8.4.3 Economic assessment

The evaluation of the economic assessment is given in Figure 27 for the two considered fU. In the realm of the LCOE, it was shown that treatment systems with a thermochemical post treatment step could compete with the base case "AD". This indicates that the secondary energy exploitation step may be economically worthwhile for treatment systems with a high energy conversion rate. For the second fU (i.e. treatment of 100 kg of raw sewage sludge), AD showed burdens that were ~30-50% lower in comparison to treatment paths with a thermochemical post-treatment step. For the first fU (i.e. production and export of 1 kWh of exergy), a high energy recovery rate could reduce the economic burdens significantly as the burdens were divided by the exported exergy amount. For the second fU, the energy recovery rate was credited according to the revenues for heat and electricity and were thereby subtracted from the economic costs. Thereof, the positive impacts of a high energy output were reduced in this case. For "AD+G" (the system with the highest energy recovery rate) this effect became most notable. For the assessment according to the LCOE, the median costs were 10% lower than those of "AD". For the mass-specific fU the treatment costs were ~55% higher than those of "AD".

When comparing the treatment systems that contained a thermochemical post-treatment step, according to the exergetic fU, the systems "AD+G" and "AD+HTC+G" showed the lowest economic burdens at median LCOE=93-95 ct/kWh. "AD+HTC+I and "AD+I" held economic burdens which were ~25-40 ct/kWh higher than the treatment path "AD+G". The costliest system was "AD+P+I", which peaked at median LCOE=1.70 ct/kWh. For the mass-specific evaluation, the economic burdens for treatment paths with a thermo-chemical post-treatment step, were similar, especially when taking the large uncertainty ranges into account. Their median values were between 1.2 and 1.4 ct/100 kg FM.

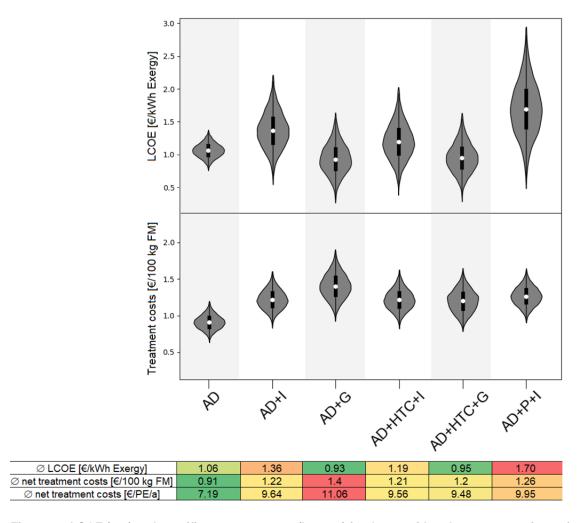


Figure 27: LCOE (top) and specific treatment costs (bottom) for the considered treatment options of sewage sludge. Violin plots represent the distribution curve. Outliers were excluded (Z-score<3). Median LCOE and treatment costs are stated in the table below the graph. The treatment costs of sewage sludge were furthermore referenced to PE/a in the last row.

In the context of sewage sludge treatment, it is worthwhile to put the economic burdens of sewage sludge treatment into context with the wastewater charges. Average (cost-covering) wastewater charges were 131 €/a/PE in 2015 (CIVITY MANAGEMENT CONSULTANTS GMBH & CO. KG 2015). The price index between 2015 and 2020 for wastewater treatment increased from 100 to 101.3, which indicated wastewater charges of 132.7 €/PE/a for 2020. Assuming that one PE represents 15.8 kg DM/a which is discharged to a WWTP (IMHOFF ET AL. 2018), the waste water charges can be put into relation with the treatment cost of sewage sludge. In Figure 27 the treatment costs for sludge according to the different product systems are given with reference to PE/a. In comparison to the base case "AD" the costs increased by 2.3-3.9 €/PE/a. The additional costs resulting from a thermal post-treatment and an individual P recovery may therefore be considered marginal. It should be noted that hereby net treatment costs for sewage sludge were referenced to gross wastewater charges and an adjustment for taxes is missing.

While a thermochemical post-treatment including P recovery puts further economic stress on the treatment for wastewater, the rough calculation on monetary costs showed that the overall treatment costs will not increase drastically. Against the given uncertainties and the varying outcomes for both fU, a clear recommendation to either treatment path is not given. By implication, this means that the economic performance of either treatment path is not an exclusion criterion, and the treatment path should be chosen according to the environmental performance. Although the treatment according to "AD" presented a cheaper option for sewage sludge treatment, recent developments signal that costs for this treatment path are constantly increasing. The gross disposal costs of sewage sludge according to an agricultural utilization increased from 200€/t DM in 2015 to 425€/t DM in 2019 (HILMER 9/11/2019), as the acceptance for spreading sewage sludge is constantly declining in the agricultural sector.

### 8.4.4 Conclusive recommendations

With the amendment of the sewage sludge ordinance significant changes in the regulation will take effect by the end of this decade. Sludge from WWTP with a capacity >50,000 PE will not be permitted to be spread on agricultural areas, while P-recovery becomes legally binding. As ~60% of all sewage sludge is treated in large-scaled WWTP (HABERKERN AND RETAMAL PUCHEU 2020), a high relevance for choosing an environmentally sound treatment paths is indicated.

The transformation process towards meeting the new regulations has already begun. In Figure 28 the development of sewage sludge treatment paths is given. The amount of sewage sludge treated in the agricultural sector dropped from ~40% to 17% during the last two decades, while at the same time the amount of sludge which was thermally treated increased from ~20% to ~75%. Thermal treatment alone, will however not be compliant with the future regulations, as P must be recovered along the treatment path. If sewage sludge is co-combusted, P must already be recovered from the wastewater or sludge. Otherwise, if P is recovered from the ash of the sewage sludge, only a mono-treatment will be feasible. Currently, mono-incineration of pre-dried sludge is the method of choice (UBA 2018).

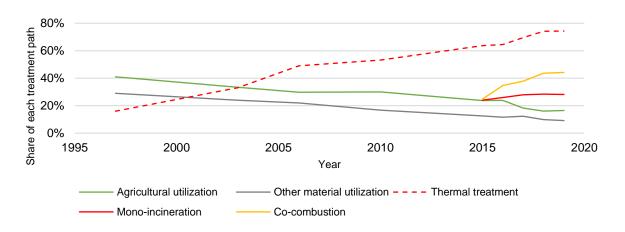


Figure 28: Shares of treatment paths for sewage sludge utilization during the last two decades (Roskosch AND HEIDECKE 2018; DESTATIS 2020c)

The environmental assessment modelled the treatment path of mono-incineration according to "AD+I" and found that this treatment path was justifiable from an environmental point of view. The implementation of an intermediate carbonization step according to "AD+HTC+I" concluded in similar impacts. The HTC process was found to reduce the heat demand significantly in comparison to drying, but the positive effects were not notable in the LCIA. This was lastly an effect of the specific setup of the treatment path. The heat for the dryer could be obtained from the co-produced heat of the incinerator, while the HTC-plant was operated decentralized and external heat (in the form of natural gas) was required. Two potential options are conceivable, where the HTC process according to "AD+HTC+I" is preferential over direct incineration according to "AD+I": (i) heat for the dryer cannot be provided by the incineration process, (ii) the distance between the WWTP and the incineration plant are far apart from each other. In the latter case, the decentralized HTC step reduced the transportation effort by the factor 2.5, as less mass was transported to the incinerator. For the calculations, 50 km between the incinerator and WWTP were assumed and at this point the environmental impacts of "AD+I" and "AD+HTC+I" broke even. Further distances would therefore tip the environmental balance in favor of the system "AD+HTC+I". Lastly, it also appears plausible to operate the HTC-plant centrally and fuel it with heat produced from the incinerator. The heat demand for a HTC unit in comparison to a drying unit is lower and in turn the energy export from the incinerator could be increased. It must however be kept in mind that the HTC process requires heat at a higher temperature level in comparison to a drying unit. With regards to electricity, it is expected that the efficiency of the incinerator would not drastically increase in comparison to "AD+I".

A high environmental performance was achieved for the system "AD+G" and "AD+HTC+G". "AD+G" is however only partially recommended. Although, the energetic performance of "AD+G" was the highest, the high demand in external energy for pre-drying the substrate is not justifiable. Therefore this treatment path should only be applied if the heat is not satisfied

by fossil resources or if the LHV is increased otherwise (e.g. mixing sewage sludge with woody biomass (RAMACHANDRAN ET AL. 2017)). "AD+HTC+G" showed the highest overall performance of all systems and is the recommended treatment path for sewage sludge. This was true for both fU that were evaluated. Also, the economic performance would not compromise this treatment path. In contrast, the implementation of two concepts that have only partly reached market maturity may be disadvantageous. By the end of 2023 operators of WWTP must provide a concept for treating sewage sludge and it appears unlikely that a treatment path with such high technological uncertainties will be chosen.

Lastly, direct spreading of sewage sludge does not remain an option for the referenced WWTP (i.e., 600,000 PE) and is only valid for small-scale treatment plants. Beneficial aspects of "AD" are the reduction of global impacts such as the GWP or the fossil depletion. Also, the economic assessment favors this treatment path. Its application is however only recommended, if it can be ensured that toxicity aspects are not compromised. The admissible maximum quantity of heavy metals that can be spread via sewage sludge on agricultural areas (i.e. limit values of heavy metals x amount of sewage sludge, which may be spread per year) appeared to be too high. Either the limit values or the admissible amount of sewage sludge, which can be spread per year, should be reduced to achieve an adequate dilution effect for heavy metals in general, and the elements copper and zinc in particular. The amendment of the Sewage Sludge Ordinance does however not envisage a deviation from the current regulations.

### 8.5 Limitations

### 8.5.1 Methodological limitations

### a. Energy models

The common denominator of both the economic and environmental evaluation was the net exergy production of the systems. Thereby the thermal and electrical energy consumption and production were contracted. For conventional treatment modules like anaerobic digestion, incineration or drying processes, empirical values were available, and it is expected that a high convergence with real operation conditions was depicted. For alternative technologies such as the HTC-plant, the gasification and the pyrolysis plant, the systems were modelled according to the principles of (i) the conservation of energy, (ii) equilibrium conditions and the (iii) principles of heat transfer. In that context it was indispensable to fall back on a set of assumptions. The highest level of uncertainty stemmed hereby from losses during heat transfer and the lack of thermodynamic state values. The modelled values were tested against literature data and showed a good conformity. However, also literature values were often based on modelling

approaches or lab-scaled experiments. This was counteracted through the incorporation of uncertainty ranges in the modelling phase.

In addition to the conservation of energy, the conservation of mass and elements was maintained during the models. The transfer coefficients and distribution factors were sourced from literature data (Electronic Annex), equilibrium models, on-site conducted lab-scaled experiments (MAYER ET AL. 2020; STOBERNACK ET AL. 2020) and in case of incineration processes from modelling tools provided by DOKA (2017). Since all waste streams and their derivatives were at some point converted into energy, erroneous results on a process level were partly compensated on a system level. If for instance the organic fraction of a substrate was not transferred to the gaseous phase in the pyrolysis process, it remained in the pyrochar. Both derivatives were eventually converted into energy and a similar overall energy output was given on a system level. Higher discrepancies in the energy balance were enabled when material was transferred to liquid material streams. For instance, when organic material was transferred to the process water during the HTC-process it evaded an energetic exploitation via the combustion of the HTC-char. Simultaneously, the module for process water treatment was subject to higher burdens. Although the retentate of the process water treatment module was finally incinerated, the net energy production for the process chain for water treatment was negative.

### b. Environmental assessment

The main limitations resulted from the determination of emissions. They did not stem from specific measurement campaigns but were rather sourced from literature values and adjusted to the waste composition or were modelled according to stoichiometric calculations and abatement factors. If the modelled emissions would exceed the statutory requirements, they were accordingly reduced. As treatment plants would have to comply with the respective regulations, only medium deviations from a "real" plant operation are expected.

Finally, also the methodology of eLCA is subject to limitations. As shown throughout the assessments, toxicity effects cannot be accurately depicted and put into context. A risk assessment would state a necessary supplemental approach. Also, many effect mechanisms are not yet fully understood or implemented in the LCIA methodology. Examples are plastic emissions to the biosphere (e.g. during the spreading of digestate) or the effect of CO<sub>2</sub>-emissions to marine ecosystems. A high uncertainty is also introduced when midpoint categories are aggregated to endpoint categories. Scientific approaches are implied to model endpoint categories, but lastly a strong subjectivity will always be introduced by the practitioner.

#### c. Economic assessment

With respect to the economic evaluation, a set of simplifications was introduced to approximate the economic burdens. They included the implementation of a constant degression exponent and the implementation of literature data that were not necessarily tailored to the considered substrates. Finally, some literature sources only stated the total investment and O+M costs rather than presenting disaggregated cost positions, which hindered a verification of the data. The aim of the economic assessment was however not to give a detailed and high-quality cost estimate, but it was rather used as a secondary decision aid, whether a treatment path would result in disproportionately high economic burdens. The implementation of uncertainty ranges, according to the deviations in the literature data increased the robustness of the model additionally.

## 8.5.2 Limitations in the application of the results

The presented results were based on an Ex-Ante approach, which implies that especially novel technology-forms were considered fully functional. In combination with the considered substrates, the technological feasibility is yet to be confirmed. Especially treatment paths that contained an HTC and/or gasification step were partly characterized by a high energy conversion rate and a high environmental performance. Whether these performances can be achieved and whether the systems are functional within industrial-scaled plants is yet to be determined. High ash contents in the substrates or their respective chars may hinder the gasification process. Hereby problems related to ash melting, accumulation or agglomeration were reported (VOGEL ET AL. 2006; QUICKER ET AL. 2017). For the HTC-process (i) the treatment of process water and (ii) the heat recovery from the suspension requires further research. The implemented concepts are based on demonstration plants (process water treatment: carbonSolutions Deutschland GmbH (STARK AND MAAS 2012), heat recovery: TerraNova (BUTTMANN 2011)) and appear to be feasible, but leave room for further improvement.

Lastly, each technology or combination of technologies was connected to a specific treatment path. The presented results are only valid in conjunction with the depicted process chains, the given assumptions, and the underlying operational parameters. In this context it is also particularly important that the presented heat management is applied. This means that heat which is produced in the systems is also utilized within the system according to the given system boundaries. If internally produced heat remains unused and the heat demand is satisfied by external fossil fuel sources, the environmental balances, especially the fossil depletion potential and GWP, would be drastically impaired. Vice versa, if external heat is derived from biogenic heat sources the environmental balance can be profoundly enhanced.

	Chapter 9
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# **Conclusion and Recommendations**

This work analyzed five main technological conversion systems for organic waste streams towards the production of energy. The approaches (i) incineration, (ii) anaerobic digestion, (iii) HTC, (iv) gasification and (v) pyrolysis were applied individually or in a cascade to the waste streams (i) waste wood, (ii) food waste, (iii) OFMSW and (iv) sewage sludge. In the process the energetic, environmental, and economic performance were determined by means of energetic modelling, eLCA and LCOE. The study aimed at finding environmentally sound treatment paths for the considered substrates, while the economic aspects were not compromised. Additionally, the meaningfulness of source segregation of these waste-streams was discussed.

As a summary the considered treatment paths are clustered in the categories A, B and C. Systems listed in category A represented the preferential treatment options, while systems in category C stated the least beneficial system. The results are given in Table 3. The evaluation is a recapitulation of the quantitative assessment and qualitative discussions, but the approach does not follow a scientific basis. Given the underlying uncertainties and the multitude of impact categories, with different relevancies, the presented categorization is a synthesis of individual discussions and comparisons between the considered systems.

Table 3: Clustering of the different treatment paths according to their environmental and economic performance. Category A=highest performance, B=medium performance, C=lowest performance.

Substrate	Sustainability aspect	category A	category B	category C
Waste wood	environmental	I, co-I, G	P+G, P+I	HTC+G, HTC+I
	economic	I, co-I, G	P+G, P+I, HTC+I	HTC+G
Food waste	environmental	AD, AD+I, AD+G, AD+HTC+I, AD+HTC+G	I, G, P+I, HTC+I	P+G, HTC+G
	economic	AD, AD+I, AD+G, AD+HTC+I, AD+HTC+G	I, G, HTC+I	HTC+G, P+G, P+I
OFMSW	environmental	I, AD+comp, G, AD+HTC+I, AD+HTC+G	HTC+I, AD+I, AD+G	HTC+G, P+I, P+G
	economic	I, AD+comp, G, HTC+I, AD+I, AD+G, AD+HTC+I, AD+HTC+G	HTC+G	P+I, P+G
Sewage sludge	environmental	AD+I, AD+HTC+I, AD+HTC+G	AD, AD+G, AD+P+I	-
	economic	AD	AD+I, AD+G, AD+HTC+I, AD+HTC+G, AD+P+I	-

In conclusion the assessment showed that conventional treatment systems are represented for all substrates in category A. Their economic performance did not state an exclusion criterion. Incineration was found to be preferential for the treatment of waste wood ("I"), anaerobic digestion followed by spreading of digestate was one of the most beneficial options for food

waste and OFMSW ("AD" and "AD+comp") and the incineration of digested sewage sludge also stated an acceptable treatment path ("AD+I"). Generally, the spreading of digestate from OFMSW and food waste is recommended, as the finite resource P can be recycled. This was true, even though the hotspot analysis proved that the post-treatment of digestate, i.e., composting (in the case of OFMSW) and spreading were the main drivers of environmental burdens in these product systems. Only if the application of organic fertilizer is not meaningful, an energetic treatment is preferential. Also, in this case, conventional treatment paths are found in category A. For OFMSW, direct incineration is eligible ("I") and for food waste a combination of anaerobic digestion and incineration ("AD+I").

The positive aspect of this résumé is that the waste management sector can fall back on technologies that reached market maturity to meet sustainability aspects according to the Circular Economy Law. On the downside this study implies that innovative treatment systems, like gasification or HTC do not hold the potential to enhance the environmental performance significantly within the given framework. The pyrolysis of organic wastes even proved to be a detrimental treatment path in the realm of WtE. Only for few constellations both the gasification and HTC were found to yield disproportionately high benefits. As both systems can be operated decentralized, they were applicable, where long transportation distances between the waste collection point and a centralized incineration plant would compromise sustainability aspects. Examples are a decentralized HTC-step of sewage sludge, prior to its transportation to a centralized incineration unit or a combination of anaerobic digestion, HTC and gasification ("AD+HTC+G") for the treatment of OFMSW.

Based on this outcome, the main recommendation towards a more sustainable waste treatment of organic waste generally points in the direction of more comprehensive flue gas treatment systems in conventional treatment systems. This implies that limit values for NO<sub>x</sub>, NH<sub>3</sub> and SO<sub>x</sub> emissions during combustion processes must be tightened to reduce the impacts in the categories particulate matter formation, acidification and ozone formation. Additionally, a limit value for N<sub>2</sub>O emissions should be introduced when waste is combusted. Its high contribution to the GWP reduces the environmental performance of incineration processes. A displacement of urea with ammonia in the SNCR and maintaining over-stoichiometric burnout zones at temperatures >850°C are effective measures to reduce N<sub>2</sub>O emissions. Finally, all facilities, where organic waste is stored or biologically treated, should be enclosed. The exhaust air ideally undergoes an acidic scrubber prior to a biofilter. This does not only reduce NH<sub>3</sub> emissions more comprehensively, but also reduces the potential for a new formation of

 $N_2O$  in biofilters. When composting units are implemented in the treatment path, the temperature level and air flow should be optimized so that  $N_2O$  and  $CH_4$  emissions are kept to a minimum.

The second research question of this assessment targeted the status of source segregation in Germany. Based on the results of this work, the currently given system of source segregation in Germany appears to be sufficient. Withholding OFMSW from mass incineration and treating it in anaerobic digestion plants is recommended. This treatment enabled the recovery of nutrients from the digestate and could thus reduce the fossil depletion potential. Additional source segregation of food waste from OFMSW was not found to result in beneficial aspects, since the preferential conventional pathway for both OFMSW and food waste was anaerobic digestion. If a market for digestate is not given, it may be worthwhile to collect food waste separately from OFMSW. The remaining part of OFMSW may be combusted directly and the food waste could undergo anaerobic digestion first, followed by dewatering (either conventional drying or HTC) and an energetic exploitation. For waste wood (AIII, AIV), source segregation should be maintained, even if it is incinerated in a municipal waste incinerator, as it constitutes a hazard-ous waste stream. Only by doing so, this waste stream can be properly monitored. For sewage sludge, the aspect of source segregation was not relevant, but, if P needs to be recovered from the ash, a mono-treatment for the thermochemical post-treatment is required.

# **Appendices**

### Electronic annex

https://th-koeln.sciebo.de/s/JqOgKoKfbCK6iGP

## Appendix A. Major sustainability aspects in the waste management sector

### a. Social aspects

Untreated and uncollected waste represents an odor nuisance and can lead to a mutilation of the natural surroundings by means of land transformation and occupation. Secondly, waste may function as a hazard to human health. A risk is either provoked by (i) short- or long-term effects of hazardous waste itself, (ii) secondary, health endangering degradation products, such as mold spores, putrid bacteria or toxic by-products or (iii) the attraction of animals, such as rodents, which may function as vectors for diseases (KAZA ET AL. 2018). The installation of a functioning waste collection system and its management at a centralized treatment site may diminish these drawbacks but could also lead to a displacement or externalization of the problematic aspects. The respective waste treatment sites are often subject to increased emissions (such as odor, noise or direct secondary human health affecting emissions resulting from the treatment of waste), which may restrict the livability of residents. A participative approach by means of stakeholder engagement throughout all stages of the management process is considered an expedient approach to achieve a high degree of public acceptance and to counteract a "not in my backyard" mentality (TAELMAN ET AL. 2018).

Next to the residential population, the social aspects of actors, which are directly involved in the waste management may be compromised. Occupational health, especially in countries with low safety standards could diminish the sustainability aspects of some waste treatment paths (Cointreau 2006). On the positive side, it may finally be argued that the waste management sector is a major employer. In Germany this sector counted for instance >170.000 employees in 2018 and is constantly growing (DESTATIS 2020b). This seeming benefit is however to be considered critically, as an increased employment rate is among others propelled by a constantly increasing waste production.

## b. Environmental aspects

Both treatment and lack of treatment may lead to versatile global and local environmental impacts. Depending on the fate of waste, resource and energy consumptions as well as emissions lead to environmental impacts. Direct emissions occur during natural degradation processes or chemical, biochemical, thermochemical or mechanical conversion steps of the respective waste. Resource depletion and indirect emissions are driven by external energy consumption and the need for capital and ancillary goods: buildings, machinery or storage facilities need to be constructed and maintained and consumables, aggregates, chemicals or short-lived packaging material are spent during waste management.

While waste streams are generally viewed as source of environmental stress, they must at the same time be considered as resource of energy, secondary material and/or nutrients. This may partially offset the environmental burdens, which are caused during waste treatment. Considering waste as a renewable resource or energy is however dangerous and should be refrained from, as it would imply that an increase of waste states a positive development. Additionally, the source-material itself is often not renewable and only for few biogenic waste streams this thought pattern may be applicable, as the waste stream itself can be replenished. However, it must be kept in mind, that fossil resources, which are not renewable may be required along the value chain.

### c. Economic aspects

Within waste treatment systems, waste is mostly free of charge and revenues may be generated by secondary products (e.g. recyclates, energy etc.). In most cases, waste treatment is still associated with net economic burdens, resulting from the waste collection and the investment and operational costs of the waste treatment. In this context economic costs are often opposed by environmental aspects. A general statement that more environmentally friendly treatment options would always result in higher economic costs cannot be given. Especially if waste management systems are ill-defined, i.e. inappropriate for the treated waste, higher monetary expenses may automatically enhance the environmental performance. However, to keep up with (i) increasing limit values, (ii) higher efficiencies and (iii) higher recycling and recovery rates that are demanded in the waste treatment sector, a higher engineering complexity is implied and therefore higher expenses arise.

# Appendix B. Waste hierarchy

The five steps outlined by the European waste hierarchy, which were formulated in the realm of the Waste Frame Work Directive, give a guideline on the waste management towards an environmentally friendly waste management and a circular economy (Directive 2008/98/EC):

- 1. Prevention. Waste prevention and reduction are the paramount objectives but are so far largely neglected. The waste production rate in Germany stayed constant over the last two decades but is high in the international comparison. Waste production rates have been increasing globally and are deemed to rise further. Generally, a strong correlation is given between the gross domestic product and the annual waste production and therefore developed countries are characterized by a particularly high waste production rate. Next to an increased consumption of goods, also tertiary purchasing motives, like convenience in consumption, which may result in an increased packaging demand led to this development.
- 2. Reuse and preparation for reuse. Any operations that change the status of a component or product from waste to its original or similar purpose are considered the second-best option. Checking, cleaning or repairing recovery actions prolong the lifetime of the product and thus indirectly reduce the amount of waste. For some waste streams, especially for organic waste, this treatment path may not be applicable.
- 3. Recycling. Within this recovery option, waste materials are partially or fully reprocessed into useful products, materials or substances. The recovery of nutrients from organic material and their return to the nutrient cycle is included in this step. Recycling rates, defined as share of the input, which enters treatment plants classified as "recycling"

plants" are particularly high in Germany for source segregated materials and products: electrical equipment (100%), glass (100%), paper and carboard (99%), biowaste (97%), packaging (90%) and other recyclables (81%) (DESTATIS 2018a). The recycling rates however only cover source segregated material and do not give any information on the recycling efficiency. For plastic waste for instance only 15.6% of the total disposed feed material is used as recyclate for plastic products (FUHR ET AL. 2019).

4. Recovery. Recovery in this context means that waste serves a useful purpose in replacing other materials, which would otherwise have fulfilled a distinct function. Among others, it comprises the recovery of metals, solvents, acids, bases or other inorganic material. It also includes the land treatment resulting in an agricultural or ecological benefit. In most cases it refers however to the energy recovery from waste. Whether a combustion system is classified as a recovery plant from a legal point of view is determined by a non-scientific energy efficiency criterion, often referred to as the R1-formula (Equation 5). A treatment plant, permitted after 31. December 2008 is defined as recovery plant if their energy efficiency (R1) exceeds 0.65.

$$R1 = \frac{E_p - (E_f + E_i)}{0.97 \cdot (E_w + E_f)}$$
 Equation 5

Ep is the annual heat and electricity production, where electricity is multiplied by the factor 2.6 and utilized heat by the factor 1.1.  $E_i$  is the yearly input of external fuel to the system for the provision of steam,  $E_i$  is the amount of annually imported energy without  $E_w$  and  $E_i$  and  $E_w$  is the annual energy content (given as lower heating value) of the waste treated in the plant.

More detail on recovery operations is given in Annex II of the Directive 2008/98/EC.

5. Disposal. Disposal is the last and least favorable step of the waste hierarchy. It describes waste incineration without energy recovery (in accordance with the R1-formula), discharge to water bodies, deep injection or final disposal to landfill sites. More detail on disposal operations is given in Annex I of the Directive 2008/98/EC.

### Appendix C. Steps of an eLCA

The conduct of an eLCA is formalized by the ISO 14040/44 by four consecutive, yet iterative steps. They are subsequently outlined.

### a. Definition of the goal and scope

Initially the target of the study must be precisely defined, and a reference unit must be chosen. The so-called "functional unit" (fU) determines a reference to which all environmental impacts are normalized to. The fU must be precisely described and all analyzed systems in the realm of an eLCA must deliver this fU. In the next step, the modelling environment must be specified. It must be determined, (i) whether an attributional or consequential approach is chosen, (ii) whether a comparative eLCA is conducted or a hotspot analysis is aimed at, (iii) which temporal and geographical scope is applicable and (iv) which allocation approach is chosen. Allocation determines, how useful co-products within a multifunctional system are handled. In the next step (v) an impact assessment method must be chosen and if environmental impact categories are to be excluded from the analysis, this needs to be stated and justified. Lastly, (vi) the

system boundary of an analyzed system must be outlined. In the scope of waste management, the evaluation can start already at the point at which material undergoes a transition from the status of a useful material to an unwanted or unusable material. According to the Waste Framework Directive the transition is complete, when "the holder discards or intends or is required to discard" a substance or object. This is considered the "cradle" of the waste material. In the realm of a life cycle view, waste management comprises all activities related to the collection, transport, pre-treatment, treatment, post-treatment and the final disposal or recycling/reusing of by- and co-products. Depending on the destination of the secondary products, the treatment options can be considered as a cradle-to-cradle or cradle-to-grave approach. Hereby, grave refers to the final disposal of a by-product (e.g. landfilling of fly ash) and cradle refers to the recycling or reuse of a secondary (useful) product, a co-product or a refurbished product. Ideally an eLCA covers the entire life cycle, but within a comparative eLCA it is legitimate to exclude processes that are the same for all systems.

This first step of the eLCA must be carefully conducted, as it determines the boundary conditions of the model. It has the potential to greatly affect the outcome of a study. The respective ISO-norms give limited guidance upon the determination of the goal and scope or, positively worded, allow for individuality and degrees of freedom. Therefore, it is hardly possible to compare quantified impacts of two studies of two different practitioners, as the boundary conditions would mostly deviate.

## b. Life cycle inventory

Within the eLCA all processes and sub-processes within the process-chain associated with the waste management must be assessed and quantified. This is referred to as Life Cycle inventory (LCI). The extend of the process chain is marked by the system boundary. Any activities beyond the system boundary are disrespected. A comprehensive inclusion of all processes is key and an abandonment from this principle is only possible in justified, exceptional cases. The entirety of all processes within the system boundary is referred to as product system. For each process, the inputs and outputs must be quantified. These so-called flows contain all material and energy flows, including ancillary goods, capital goods and emissions. In most cases, they lead back to other process and thus the interlinking of all process, connected to a product system, eventually leads to a complex network (e.g. the provision of heat by natural gas leads back to the production of a boiler for this purpose and the natural gas production, which themselves have predecessors etc.). Due to the availability of powerful and comprehensive databases that partly feature more than 10,000 process, generic process must not be individually modelled anymore.

### c. Life cycle impact assessment

The inventoried in- and output flows are finally transferred into environmental impacts. Within eLCA a broad number of impact categories are assessed. They refer to resource consumption (e.g., fossil or mineral resource depletion), human health (e.g. particulate matter formation, toxicity or ozone depletion) and the ecosystem quality (e.g. eutrophication, acidification or climate change). Each of the individual environmental impacts contains a reference unit (e.g. Climate Change is measured as kg  $CO_{2,Eq}$ ) and emissions or resource consumptions are translated into impacts by means of an impact factor.

### d. Interpretation

All of the previous steps are subject to constant interpretation. It must be validated, whether the scope of the study would always result in the desired goal, whether the LCI is comprehensive for each process and whether all processes relevant for the product system are included. Finally, the results of the LCIA must be interpreted. Initially it should be ensured that results are calculated in the right order of magnitude, by comparing it to similar product systems that have been previously validated, e.g., product systems that are already part of a professional database. Afterwards hotspots within the product systems should be identified and on this basis a resilient data situation must be prepared for flows that result in major impacts. Subsequently, relevant impact categories for the considered product systems must be chosen. When impacts within an impact category result primarily from the background system, this indicates that they are of reduced relevancy. Also, the aggregation of midpoint categories to endpoint categories (areas of protection) can aid in the interpretation. Midpoint categories that contribute little to the respective endpoint categories tend to be of minor importance. This methodology is referred to as normalization and weighting. As it is both based on subjective decisions and on a non-scientific approach, it must be applied cautiously.

Finally, the robustness of the model can be elevated by including an uncertainty assessment. Huijbregts (1998) identified five sources of uncertainty and variability during the conduct of eLCA:

- Parameter uncertainty: imprecise, incomplete or lack of data with regards to the inventoried data and the impact factors.
- Model uncertainty: some aspects within the eLCA modeling are overly simplified, e.g. a linear response to emissions is assumed in environmental systems or impact factors are derived from simple models.
- *Uncertainty due to choices:* choices with respect to the modelling environment are unavoidable and may result in large deviations.
- Spatial variability: the response of emissions to ecosystems may vary between different locations.
- Temporal variability: daily, monthly or yearly variations of weather conditions such as temperature or wind speed would affect the impact of certain emissions. Also, the chosen time horizon in the LCIA can alter the results.

Within uncertainty analyses, the first aspect, i.e., parameter uncertainty is given the highest priority. Sensitivity analysis or Monte-Carlo simulations (HEIJUNGS 2020) are apt tools to conduct an uncertainty analysis.

# Appendix D. History of waste management in Germany

Unsanitary landfills, open burning or missing waste collection systems reflect situations that are present in many low-income countries today. A similar situation was given in Germany in the previous century (BUCHERT ET AL. 2013). Although, local attempts towards a modern waste management were already made by the end of the 19<sup>th</sup> century – e.g., installation of the first waste incinerator in Hamburg (1893) or the introduction of compulsory waste bins in Berlin (1895) – they were not introduced nationwide. The convergence to an environmentally and socially compatible waste management solution started only in the 1960s. Economic incentives, legal regulations and a gradual change in the mindset of the society, induced by awareness raising led to the present waste management system.

In the 1960s only 37% of the municipal solid waste were treated at sanitary landfills (~130 units), composting plants (16 units) or incineration plant (30 units) (BILITEWSKI AND HÄRDTLE 2013). A majority of the accruing solid waste was disposed of at ~50.000 unsanitary landfills (KLOEPFER ET AL. 1994). At this time landfill sites were solely subject to a reporting obligation but were not governed by requirements and thereof contained no protection measures against the environment or the local population. Legal bases were, if applicable, municipal bylaws. This problematic issue was first targeted in the scope of the protection of water bodies. Issued in 1957 and incepted in 1960 the *Water Resources Act*, which ensured that waste material was deposited in a manner that groundwater contamination did not occur (SMEDDINCK AND KLUG 2016). Further regulations on waste management and treatment were embedded in the Trade Regulation and the *Federal Pest Control Act*.

Finally, in 1972 the *Waste Disposal Law* was enacted. For the first time, a nationwide law that was explicitly dedicated to waste management was present. As implied by the title, it was primarily focused on waste disposal. Recovery or recycling from waste was initially disregarded. In connection with this law, (i) smaller unsanitary landfills were shut down, (ii) landfill sites were centralized, controlled and certain environmental standards had to be met and (iii) the responsibilities for waste collection and treatment were clearly allocated. Infringements were punishable. In 1974 the *Emission Control Act* came into force and emissions resulting from the waste management were further mitigated. As response to previous efforts made by member states, the European Community issued the *Waste Framework Directive* in 1975 (FARMER 2012). The maxims of the waste hierarchy were therein incorporated. Member states, including West Germany, were obliged to take measures to avoid or reduce waste, while minimizing the impacts on human health and the environment. Secondly, resources in waste streams should be recycled or recovered. By means of the *German Waste Management Program* of 1975 these objectives were formalized but were not legally binding. A voluntary implementation on the municipal level was envisaged but did not take place.

Between 1976 and 1986 the Waste Disposal Law was amended four times and was finally renamed into Waste Law (4th amendment). A commandment towards waste prevention and utilization became an inherent part of the Waste Law and marks a transition from a sole disposal to recovery of waste fractions (BMU 2018). Simultaneously, (i) a labelling obligation of products with a high pollutant content, (ii) an obligation for a separate disposal of waste with a high pollutant content and (iii) the obligation for a mandatory deposit of certain products (§ 14 AbfG) were adopted. The government was thereby entitled to make producers liable of an environmentally friendly post-treatment of their products at the end of their lifetime (KRANERT AND CORD-LANDWEHR 2010). Trade and industry associations agreed on voluntary commitments towards the collection, withdrawal and recovery from certain waste streams such as waste paper, glass, batteries or end-of-life vehicles. In 1996 the Circular Economy and Waste Law replaced the Waste Law. The objectives of an environmentally friendly waste management according to the waste hierarchy (the hierarchy is detailed in 1.1) and the concept of a product responsibility were consequently pursued. In the realm of the amendment of the European Waste Framework Directive in 2012 the Circular Economy Law was updated to comply with the directive. Major changes were not implemented.

To the present day, the *Circular Economy Law* paved the way for 19 legislative degrees (some were partly annulled again or transposed into laws). They concretize the treatment paths for individual waste streams, e.g. sewage sludge, biowaste, waste wood etc., determine product

and production related operations and give the requirements for waste disposal. The *Circular Economy Law* indirectly aided in a landfill ban of untreated material that exceeds an ignition loss of 5% (effective since 2005). Furthermore, a source segregated collection of glass, paper, metal and plastics became part of the waste management strategy in Germany. Since 2015 all private households are obligated to source segregate their waste. Municipalities determine the boundary conditions for the waste separation. One of the more recent developments describes the legal regulation for the provision of separate waste bins for the organic fraction of municipal solid waste (OFMSW). It took effect only in 2015.

The preceding description on the history of waste management, suggests that a top-down approach was followed to establish today's waste management system. In reality, four actors of the waste management constantly interact(ed) (LÄPPLE 2009):

- 1. Legislators and executive organs: the European Union sets the legislative framework, which is harmonized on the national level. As a rule, the German waste law exceeds the requirements of the European regulations on waste management. On the federal state level, implementation laws are designed in accordance with the national law and administrative, financial, executive and planning task are regulated. On a municipal level, national and provincial state law is executed in the realm of waste statutes and waste management concepts. Waste management fees are defined at this level. (KRANERT AND CORD-LANDWEHR 2010)
- Economy: The economy generates and distributes products. In connection with the
  product responsibility for the entire lifecycle of a product, the economy is directly involved in the waste management process. Additionally, private-sector companies operate waste management services. They are in direct competition to municipal service
  providers.
- Consumer: Citizens act as consumers of economic goods and producers of waste.
   Their consumption behavior has a direct impact on the waste generation. Their sorting accuracy at the end of the life of products affects the grade purity of recyclable material.
- 4. Public: citizens and citizens' initiatives can directly influence the waste management sector. Cooperation or resistance towards projects or regulations in the waste management sector is an appropriate approach and was previously utilized. E.g. during the 1980s incineration plants were impeded by the local population and emission limits were severely tightened to improve the acceptance for new plants. Participation, information campaigns (e.g. based on scientific findings) and the filing of complaints are further instruments of the public.

By means of the efforts of all four actors, waste management underwent significant changes and was subject to major improvements throughout the last decades in Germany. To keep pace with these developments, technological setups were constantly improved (e.g. improvement of the flue gas treatment in incinerators) and technologies were newly invented or customized to treat waste material (e.g. treatment of organic waste by means of anaerobic digesters).

One actor was of crucial importance for the advancement in the waste management sector: the scientist. Over the years, numerous technologies were developed for the preparation of reuse, recycling and recovery from waste. Starting at lab-scaled size, technologies were streamlined towards the ideal configuration and finally implemented at an industrial scale. While the scientific branch played a major role in the advancement of technologic solutions, it

is today facing an additional challenge. It needs to evaluate these technologies and quantify which technology, "deliver[s] the best overall environmental outcome" for different waste streams, while life cycle thinking is adopted (Article 4 (2) Directive 2008/98/EC).

## Appendix E. Short description of the considered treatment technologies

### a. Incineration

Incineration describes a direct thermo-chemical conversion by means of oxidation. It is an over-stoichiometric combustion process and the air-fuel ratio (λ) in the total system is ≥1. The combustible parts are oxidized and transferred into the gas phase. This is accompanied by an exothermic reaction and heat is released. Exothermic reactions are opposed by the endothermic heating and evaporation of water in the fuel. During incineration five stages may be differed in the combustion chamber: (i) drying by increasing the temperature to >100°C, (ii) degassing in a reduced atmosphere by further increasing the temperature of the substrate to >250°C, (iii) pyrolysis and gasification of the organic fraction (iv) oxidation of the combustible gases generally between 800 and 1450°C, (v) post-combustion towards a complete oxidation of the previously unburned material (EC 2006; BILITEWSKI AND HÄRDTLE 2013). Depending on the technological setup, the phases are (partly) spatially separated.

Virtually all incinerators for municipal solid waste in Germany are designed as grate-incinerators (UBA 2018). They place low demands on the fuel quality (ARAFAT AND JIJAKLI 2013) and waste must, as a rule, not be pretreated. Increased moisture contents reduce their performance or lead to a standstill of the combustion process. RAND ET AL. (2000) state that the lower heating value (LHV) of the treated waste should be >6 MJ/kg throughout the seasons to enable an autothermal combustion process.

A more recent technology in this field is the fluidized bed incineration. Thereby primary air flows through loose bulk material (such as sand, ash or limestone) and a fluidized bed is formed (THOMÉ-KOZMIENSKY AND BECKMANN 2013). Currently it is widely applied for the treatment of sewage sludge and refuse derived fuel (RDF) (FLAMME ET AL. 2019). The main advantage are high heat transfer rates and the treatment of waste with poor fuel properties (high ash and moisture contents). An autothermal combustion of substrate at an LHV<4.5 MJ becomes possible (CHEN AND CHRISTENSEN 2010; FRIEDRICH ET AL. 2013). Higher monetary costs increased parasitic electricity demands and more erosion in the combustion chamber represent disadvantages of this technology. Further technologies, but irrelevant for this work, are rotary kilns (for hazardous waste) and various types of furnaces (e.g., multi-deck ovens, cyclone converter furnaces, shaft furnaces etc.).

Regardless of the technology, incineration results in the production of a hot flue gas. According to the principle of energy recovery, the thermal energy is utilized for steam production. In most cases incineration units, are designed as cogeneration units in Germany (UBA 2018). Within a Rankin cycle, superheated steam is converted within a single-stage turbine into electricity. The condensation heat can be decoupled for district heating purposes or to pre-dry substrate (e.g., sewage sludge). The export of both heat and electricity lead to a high exploitation rate of the inherent energy of the substrate and allow for a compliance with the R1 criterion.

Subsequently to the transfer of sensible heat to the superheater, flue gas needs to undergo several gas-cleaning stages to reduce gaseous and particulate pollutants. Particulate removal

is realized by means of filters and/or cyclones. Adsorptive and/or absorptive processes remove acidic gases in the realm of a dry, quasi-dry or wet flue gas treatment. Finally, the amount of nitrogen oxides is diminished by selective catalytic reduction (SCR) and/or selective non-catalytic reduction (SNCR).

### b. Anaerobic digestion

Anaerobic digestion describes a bio-chemical conversion of biogenic substrates within an oxygen-free environment. Four steps can be distinguished on a macro-level. At first, (i) fermentative bacteria convert easily degradable polymers, such as proteins, lipids and carbohydrates into amino acids, fatty acidy and sugar. This is referred to as hydrolysis. These intermediate products are then converted during the acidogenesis into organic acids and alcohols. During the next two phases, the acetogenesis and methanogenesis, organic acids and alcohols are further metabolized into methane and carbon dioxide. Acetogenic and methanogenic bacteria, involved in the last two steps mutually influence each other and therefore, these two steps take place at the same time. (BILITEWSKI AND HÄRDTLE 2013; ROSENWINKEL ET AL. 2015). The reaction network for anaerobic digestion is complex and beyond the scope of this work.

Fundamental prerequisites for a functioning anaerobic digestion process are apt biogenic substrates and milieu conditions. Complex macromolecules, such as cellulosic fibers and lignocellulose do not undergo anaerobic digestion. Toxic components within the biomass or byproduct produced during the digestion may inhibit the biological activity of bacteria (KALTSCHMITT ET AL. 2016). The ideal milieu conditions with regards to temperature, pH-value or retention time for a specific substrate can mostly only be determined experimentally. In reality, however WtE biogas plants are optimized towards a cost-efficient operation, which mostly counteracts an optimal temperature range and hydraulic retention time.

The anaerobic digestion results in a high calorific gas, namely biogas. It essentially consists of 50-70% methane and 30-50% carbon dioxide. Smaller amounts of H<sub>2</sub>S, NH<sub>3</sub> and H<sub>2</sub> may be present in the biogas. A removal or reduction of H<sub>2</sub>S is recommended to reduce the corrosion of metal parts and to counteract a poisoning of catalytic converters. Within a downstream process, biogas can either be combusted directly in a CHP plant (for most applications an Otto-Gas engine is utilized in Germany (DBFZ 2012)) or its fuel quality can be upgraded to inject it into the gas grid or to use it as vehicle fuel (SUN ET AL. 2015).

Next to biogas, a digestion residue, the so-called digestate exits the biogas plant. It consists of material that was not degraded during the digestion process. High nutrient contents make it an attractive organic fertilizer. If the contamination levels allow for it, digestate is often spread on agricultural areas. An intermediate hygienization step (e.g. pasteurization, limestone application or composting) becomes often necessary, to provide a hygienically acceptable product. A downstream utilization as energy carrier is also possible. The fuel properties of digestate are however poor (increased shares of ash, nitrogen and sulfur and a low heating value).

## c. Hydrothermal carbonization

HTC is a thermo-chemical conversion process, that leads to a mild carbonization of biogenic material. In the realm of a wet milieu, elevated temperature ranges (180°C-250°C) and pressure ranges (i.e. the respective saturated steam pressure) the substrate is converted into a lignite-like product, the so-called HTC-char or hydrochar (AXEL FUNKE 2010). In comparison to the input substrate the HTC-char is usually characterized by a higher heating value, due to a shift in the composition. Both the molar H/C and O/C ratios become narrower. The ratios serve

as one of the main quality indicators for hydrochar, where a narrower ratio indicates a more complete carbonization process. Both, longer retention times and higher temperatures in the reactor increase the reaction intensity and therefore the carbonization process.

The chemical conversion in the HTC process is mainly driven by hydrolysis, dehydration, and decarboxylation. To a smaller extend, also condensation, polymerization and aromatization take place. (REZA ET AL. 2014b). During these reactions organic material is transferred to the gaseous phase. Process gas, which is released during the HTC is essentially composed of carbon dioxide (>95%) and to a much smaller part of carbon monoxide and methane. On a trace level, volatile organic compounds (VOC) may be detected that can be allocated to the substance classes of benzenes, phenols, furans, and ketones (BECKER ET AL. 2013).

After the HTC-process, a mechanical dewatering step can separate the suspension, which is exiting the reactor. As the solid product, develops hydrophobic properties during the HTC, a high degree of dewaterability is allowed for. To enhance storability and transportability, hydrochar usually undergoes a downstream drying and pelletizing process. HTC char states a secondary energy carrier which can be further utilized. Alternatively, it was reported to be an apt soil conditioner (DAI ET AL. 2015; PUCCINI ET AL. 2018; REZA ET AL. 2014a).

The process water usually states a by-product, which requires further treatment. A larger share of organic compounds (e.g. phenols, carbonic acids, furfurals etc.) and inorganic material is normally dissolved in the process water. The chemical oxygen demand (COD) is usually used to determine the degree of organic contamination. The COD of the HTC-process water usually lies in the range between 10-80 g O<sub>2</sub>/I (BERGE ET AL. 2011; BLÖHSE 2017; MERZARI ET AL. 2019). To comply with legal regulations (e.g. a discharge to WWTP requires a COD<200 mg/I), a post treatment step is essential. Several treatment approaches have been proposed to reduce the contamination level. They are summarized in Table A. 1.

Table A. 1: Potential treatment paths for process water from the HTC-process according to STOBERNACK ET AL. (2020).

Treatment	Description				
option					
Treatment in	Discharge to waste water treatment plants is possible, but the German				
wastewater	regulation limits the waste water contamination to 200 mg/l COD (BMJV 2004).				
treatment plants	Extensive dilution or pre-treatment becomes necessary.				
Recirculation Recirculation is an apt approach to reduce the amount of process					
	generally accompanied by positive side effects: slightly increased mass and				
	energy yields may be observed in the product (KAMBO ET AL. 2018; STEMANN ET				
	AL. 2013b; WEINER ET AL. 2014). The fraction of recirculated water often				
	depends on the input materials. Materials with a high initial water content				
	decrease the amount of water that can be recirculated.				
Fermentation	The high share of organic matter in the HTC water allows for a valorization				
	through anaerobic digestion. Chemical oxygen demand (COD) ratios as high				
	as 10 kg/m³/day could be successfully treated (KÜHNI ET AL. 2015). Dilution may				
	be required for an enhanced treatment. Elimination rates of the COD were				
	found to lie in a range of ~50-75% for HTC process water (WIRTH ET AL. 2015;				
	FETTIG ET AL. 2013; BLÖHSE 2017; ARAGÓN-BRICEÑO ET AL. 2017). Methane				
	yields in a range of ~0-500 ml/g COD are postulated (MERZARI ET AL. 2019).				
	Process water may be co-fermented in an anaerobic digestion plant. However,				
	high chemical oxygen demand (COD) ratios in the plant may inhibit the				

	digestion (FETTIG ET AL. 2013; KÜHNI ET AL. 2015). In KÜHNI ET AL. (2015), phenols and other intermediate products of the HTC process are stated as inhibitors of an organic decomposition. Additionally, the COD after anaerobic digestion would, still exceed the discharge limits to a wastewater treatment plant.
Filtration	Filtration techniques such as nanofiltration, ultrafiltration or reverse osmosis may lead to an effective treatment of the process water. Large-scale applications for the treatment of HTC water have yet to be tested.
Wet oxidation	Wet oxidation removes organic compounds in the liquid phase. An oxidant such as oxygen or air reacts with the organic compounds to produce carbon dioxide and water (BHARGAVA ET AL. 2006). Within wet oxidation, elimination rates up to 70% of the COD can be found (WEINER ET AL. 2018; RIEDEL ET AL. 2015). Additionally, wet oxidation may improve the biodegradability of wastewater components (RIEDEL ET AL. 2015). Similar to the fermentation process, further treatment is required after the wet oxidation process, to meet the requirement for the discharge to wastewater treatment plants

#### d. Slow pyrolysis

Pyrolysis describes a thermochemical conversion process, where  $\lambda$ =0 and temperature ranges are between 200-1100°C. Both the retention time and the reaction temperature define, whether a pyrolytic decomposition is defined as slow (long retention time and low temperatures), intermediate or fast pyrolysis (very short retention times and higher temperatures). The transition between the three types is however fluid. For any type of pyrolysis, the products are (i) a solid, carbonized product, the so-called pyrochar, (ii) a low-calorific gaseous fraction, referred to as pyrolysis gas and (iii) the pyrolysis oil, which consists of a watery and heavy-oil fraction. The composition and distribution vary with the process management and the treated substrate. During the slow pyrolysis, the mass fractions are about equally distributed on the solid, gaseous and liquid fraction. For the fast pyrolysis the share of pyrolysis oil is as high as 75% (BRIDGWATER 2018).

Pyrolysis char is characterized by a very narrow H/C and O/C ratio, implying a thorough carbonization process. Next to a high share of carbon, pyrolysis char also retains most of the inert fraction. Therefore, pyrolysis char was previously disposed of, if the heavy metal concentration was too high (BANK 2007). Pyrolysis gas is a mixture of carbon monoxide, methane, carbon dioxide, hydrogen and to a small extend long-chain hydrocarbons. Due to an increased share of CO<sub>2</sub> and CO, its energetic value is usually low. Pyrolysis oil is a complex mixture of oxygenated hydrocarbons. The liquid fraction also contains considerable amounts of water which stem from the moisture content of the substrate and reaction water, which is formed during pyrolysis.

Subsequent to the pyrolysis process, the three products can be fractionated and used individually for an energetic or material exploitation. For the slow/intermediate pyrolysis an alternative treatment option is available. The hot pyrolysis gas and the pyrolysis oil can be combusted jointly on-site to provide heat for the pyrolysis process and/or to drive a cycle process. The pyrolysis plant must then feature a gas cleaning system similar to that of incineration units to treat the flue gas. The pyrolysis char is taken out of the system and utilized separately.

#### e. Gasification

During gasification, biogenic substrates are thermo-chemically degraded at a sub-stoichiometric oxygen availability ( $\lambda$ <1) and at elevated temperatures ( $\sim$ 700-1400°C). In contrast to the incineration process, only the pyrolytic decomposition and gasification of the substrate take place. The utilization of the gaseous product is spatially separated. Within the literature the released gas is referred to as product gas, low calorific value gas or syngas. It is composed of methane, hydrogen, carbon monoxide, carbon dioxide. Additionally (depending on the process control), larger shares of nitrogen and water vapor may be present. The gas is also subject to particulate and tar contaminants and contains acidic components such as sulfur and chlorine species.

Gasification plants exist in different variations. They can be distinguished by the reactor design and the gasification agent. The cheapest and most practical gasification agent is air, but, due to its high share of nitrogen, it leads to a strong dilution of the product gas (ZHANG ET AL. 2019). Alternatively, a mixture of oxygen and steam can be used, which however results in higher monetary expenses (KALTSCHMITT ET AL. 2016). Lastly, also steam can be used as gasification agent, but an autothermal gasification process is in this case not given. The different reactor types can be allocated to the classes "fixed bed", "fluidized bed" and "entrained flow reactors". Each system holds specific advantages and disadvantages, which are not discussed here. A comprehensive overview is given in WATSON ET AL. (2018).

In this work, two reactor types become relevant:

- Downdraft fixed bed gasifiers are apt reactor types for lumpy biomaterials with low shares of fines, such as wood or compacted char. They are usually constructed for the lower power range (< 5 MW<sub>LHV,substrate</sub>). In this reactor, the gasification agent and the biomass move concurrently in the same direction. Several zones are formed within the reactor (heating zone, pyrolytic decomposition zone, oxidation zone and reduction zone). Major disadvantages arrive from high heat losses, as the product gas exits the reactor at high temperatures. Furthermore, the water content must be <20 %, to maintain an even heat distribution within the reactor. (KALTSCHMITT ET AL. 2016; VOGEL ET AL. 2006)</p>
- Fluidized bed gasifiers are used for the gasification of fine material (particle size: max 50 mm). Within the gasifier the biosolids are mixed with the bed material (e.g. quartz sand), which allows for a fast and homogenous heat transfer. The gasification agent enters the reactor from the bottom and thus forms a fluidized bed. In contrast to fixed bed gasifiers, no zones are formed and all gasification reactions take place at the same time. Fluidized bed gasifiers are usually conceptualized for high power ranges (several MW<sub>LHV,substrate</sub>). Similarly, to fixed bed gasifiers, the product gas exits the reactor at high temperatures, which reduces the overall efficiency of the process.

Prior to further utilization, tars must be removed from the product gas. Catalytic, thermal or physical removal are the three main options for the removal of tars. Further gas cleaning, including the removal of particulates, acidic components and nitrogen species is conducted prior or post the gas utilization, depending on the utilization path. Gas cleaning systems are similar to those of incineration units. The product gas can be further processed to bio-fuels (e.g. via a Fisher-Tropsch synthesis or an alcohol synthesis) or it can be exploited for heat and/or electricity production.

Within this work, a combustion within an internal combustion engine is pursued. The generation of both heat and power states one of the most efficient transfer options to useful energy. Initially a much lower heating value of the product-gas in comparison to natural gas or biogas appears disadvantageous. During the combustion however, a proportionally lower share of combustion air must be added to the product gas. Thereof, the heating value within the combustion chamber converges with that of a high calorific gas (KALTSCHMITT ET AL. 2016). In the realm of an optimized plant model, equal efficiencies in comparison to a biogas-fueled CHP are expected for a product-gas fueled CHP plant (VOGEL ET AL. 2006).

# Appendix F. Waste collection and transport

For the initial waste collection of OFMSW and food waste a house-to-house collection was assumed. A stop-and-go transportation mode was considered and the collection route was set to 5 km (WERNET ET AL. 2016). Waste wood is typically collected at larger quantities and at fewer locations. The value was set to 1 km. The transport to the first treatment plant was conservatively estimated to be 40 km (SCHWARZ ET AL. 2015) for all treatment plants and solid substrates. The transport to a centralized incineration plant was assumed to be 20-60 km (average=40 km). The transportation to a mono-incineration plant for the treatment of sewage sludge was set to 0-100 km (average:50 km).

The transport to a lignite power plant was manually calculated. The average distance from any point in Germany to the closest power plant was hereby computed. Therefore, Germany was subdivided in a 10 km x 10 km grid in ArcGis 10.3 (Figure A. 1) and the distance to the lignite power plants was calculated via a VBA script in Excel, which was connected to the Bing Maps API. The distances were weighted according to the population. A distance between 0-612 km was found and the average weighted distance was ~200 km.

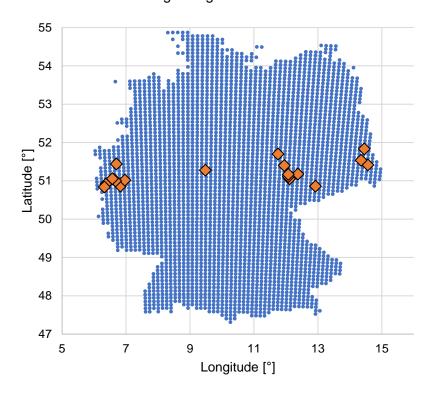


Figure A. 1: 10x10km grid points (blue dots) and location of lignite power plants (orange diamonds)

# Appendix G.Advantage and disadvantages of an input and output based functional unit

The choice in an input or output based fU may have several advantages or disadvantages.

- Within a comprehensive input-based assessment, useful energy must be credited, as it would offset environmental and economic burdens. The type of energy that is offset, can have a large impact. For instance, a large variation in the environmental and economic burdens is provoked, if electricity from lignite power plants was offset, instead of electricity from the German electricity mix. Additionally, a temporal component is involved (grid mixes change over time). The electricity price is subject to economic fluctuations and the environmental burdens resulting from the electricity grid mix varies with the technological advancement and the utilized energy carriers. Within an output-based approach, this problematic issue is not given.
- When an output-based approach is chosen, WtE options cannot be compared to other treatment options that do not produce energy along their lifecycle and only focus on sole material recovery.
- Especially for the economic assessment, operators of waste management plants are more interested in the treatment costs rather than the marginal cost per unit of energy. This would favor an output-based approach.
- By using an output-based approach, the conversion efficiency of waste to energy within
  a system receives greater recognition than it does in an output-based approach. The
  burdens, which result in a treatment system are divided by the common denominator
  energy. For WtE systems with a high specific energy output, the environmental and
  economic burdens become accordingly low. Within an input-based approach the energy, which is produced in the system is deducted as credit and the influence of produced energy in a system is reduced.
- Finally, energy is delivered in different forms, such as thermal, electrical or chemically bound energy. To maintain comparability between the systems the energy forms must be merged. However, a uniform and consistent approach does not exist in the literature.

# Appendix H. Description of the considered treatment path

lable	A. 2: Description of		ependence of the treated substrate	0.540/4/	O service and a large
		Waste wood	Food waste	OFMSW	Sewage sludge
" <b>[</b> "	Incineration	<ul> <li>incineration in a municipal waste incinerator without any further treatment.</li> <li>solidification of fly ash and landfilling of residues.</li> </ul>	<ul> <li>incineration in a municipal waste incinerator without any further treatment.</li> <li>solidification of fly ash and landfilling of residues.</li> </ul>	<ul> <li>incineration in a municipal waste incinerator without any further treatment</li> <li>solidification of fly ash and landfilling of residues</li> </ul>	N/A
" AD (+ comp)"	Anaerobic Digestion (+ composting)	N/A	<ul> <li>anaerobic digestion in a single-stage, mesophilic (37° C), wet fermentation unit.</li> <li>DM in reactor=12 wt%.</li> <li>retention time=20-25 days.</li> <li>biogas combustion in CHP units (1 onsite-CHP for onsite heat provision (200 kW) and 1 satellite plant (200 kW) for heat export to district heating system).</li> <li>pasteurization of digestate (6 wt% DM) at 71 °C and 30 minutes.</li> <li>spreading of digestate on agricultural areas.</li> </ul>	<ul> <li>anaerobic digestion in a single-stage, mesophilic (37° C), dry fermentation unit.</li> <li>DM in the reactor=28 wt%; Retention time=20-25 days.</li> <li>biogas combustion in CHP units (1 onsite-CHP for onsite heat provision (500 kW) and 2 satellite plants (2x500 kW) for heat export to district heating system).</li> <li>solid-liquid separation of digestate.</li> <li>solid part (32 wt% DM) to composting unit, liquid digestate (10 wt% DM) to pasteurization unit (71 °C, 30 minutes), remainder (2 wt% DM) to WWTP.</li> <li>composting in enclosed tunnel composting plant with exhaust air purification (acidic scrubber + biofilter).</li> <li>spreading of solid and liquid digestate on agricultural areas.</li> </ul>	<ul> <li>bustion in CHP plants (2x500 kW) to provide heat for digestion tower.</li> <li>Digestate is dewatered to 25%, remainder is sent back to WWTP.</li> <li>Spreading of solid digestate on agricultural areas.</li> </ul>

	Waste wood	Food waste	OFMSW	Sewage sludge
្ម Gasification	<ul> <li>Shredding of wood.</li> <li>Gasification in a fixed bed gasifier (~950°C).</li> <li>Syngas utilization in satellite CHP plants (3x500 kW) for heat export to district heating system.</li> <li>Solidification of fly ash and landfilling of residues.</li> </ul>	<ul> <li>Pre-drying to &lt;15% water content.</li> <li>Pelletizing.</li> <li>Gasification in a fixed bed gasifier (~950°C).</li> <li>Syngas utilization in onsite CHP plant (500 kW) for internal heat provision (drying).</li> <li>Solidification of fly ash and landfilling of residues.</li> </ul>	<ul> <li>Pre-drying to &lt;30% water content.</li> <li>Gasification in a fluidized bed gasifier (~800°C).</li> <li>Syngas utilization in onsite CHP plant (4x500 kW) for internal heat provision (drying).</li> <li>Landfilling of residues.</li> </ul>	N/A
Pyrolysis + Gasification	<ul> <li>Slow pyrolysis (500°C, 1 h).</li> <li>Combustion of pyrolysis-gas (incl. oil) in combustion chamber.</li> <li>Utilization of heat for maintaining the pyrolysis process.</li> <li>Remaining heat is used to drive a steam turbine; condensation heat of cycle process is not utilized.</li> <li>Pelletizing of pyrochar</li> <li>Char gasification in fixed bed gasifier (~950°C).</li> <li>Syngas utilization in a satellite CHP-plant (500 kW), heat export to district heating system.</li> <li>Solidification of fly ash and landfilling of residues.</li> </ul>	<ul> <li>Pre-drying to 20 % water content.</li> <li>slow pyrolysis (~500°C, 1 h)</li> <li>Pyrolysis-gas (incl. oil) combustion in combustion chamber.</li> <li>Utilization of combustion heat for maintaining the pyrolysis process and for dryer.</li> <li>Pelletizing of pyrochar.</li> <li>Char gasification in a fixed bed gasifier (~950°C).</li> <li>Syngas utilization in a satellite CHP-plant (200 kW), heat export to district heating system.</li> <li>Solidification of fly ash and landfilling of residues.</li> </ul>	<ul> <li>Pre-drying to 22.8 % water content.</li> <li>Slow pyrolysis (~500°C, 1 h)</li> <li>Combustion of pyrolysis-gas (incl. oil) in combustion chamber.</li> <li>Utilization of combustion heat for maintaining the pyrolysis process and for dryer</li> <li>Pelletizing of pyrochar.</li> <li>Char gasification in a fixed bed gasifier (~950°C).</li> <li>Syngas utilization in a satellite CHP-plant (2x500 kW), heat export to district heating system.</li> <li>Solidification of fly ash and landfilling of residues.</li> </ul>	N/A

	Waste wood	Food waste	OFMSW	Sewage sludge
Pyrolysis + Incineration	<ul> <li>Analogous to treatment path "P+G", but char combustion in centralized lignite power plant (electricity production only).</li> <li>Solidification of fly ash and landfilling of residues.</li> </ul>	<ul> <li>Analogous to treatment path "P+G", but char com- bustion in centralized lignite power plant (electricity pro- duction only).</li> <li>Solidification of fly ash and landfilling of residues.</li> </ul>	<ul> <li>Analogous to treatment path "P+G", but char combustion in centralized lignite power plant (electricity production only).</li> <li>Solidification of fly ash and landfilling of residues.</li> </ul>	N/A
HTC + Incineration	<ul> <li>Pre-mixing to reach a biomass:water ratio of 0.15.</li> <li>HTC of suspension (220°C, 3 h).</li> <li>Dewatering of suspension to ~60 wt% DM-content</li> <li>Solid part is dried from 60 wt% to 90 wt% and is pelletized.</li> <li>Char is transported to and combusted in a centralized lignite power plant.</li> <li>Solidification of fly ash and landfilling of residues.</li> <li>Liquid part undergoes a cascade of nanofiltration and reverse osmosis.</li> <li>Retentate is either recirculated or sent to an incineration unit.</li> </ul>	<ul> <li>Pre-mixing to reach a biomass:water ratio of 0.15.</li> <li>HTC of suspension (200°C, 3 h).</li> <li>Dewatering of suspension to ~60 wt% DM-content</li> <li>Solid part is dried from 60 wt% to 90 wt% and is pelletized.</li> <li>Char is transported to and combusted in a centralized lignite power plant.</li> <li>Solidification of fly ash and landfilling of residues.</li> <li>Liquid part undergoes a cascade of nanofiltration and reverse osmosis.</li> <li>Retentate is either recirculated or sent to an incineration unit.</li> </ul>	Pre-mixing to reach a bio- mass:water ratio of 0.15.	N/A

		Waste wood	Food waste	OFMSW	Sewage sludge
" AD+I"	Anaer. Dig. + Incineration	N/A	<ul> <li>Analogous to treatment path "AD", but digestate is separated into a solid (DM=24 wt%) and liquid fraction (DM=6 wt%) and both CHP are located on-site for providing heat for drying.</li> <li>Solid fraction is pre-dried onsite to 90 wt% and is transported to and combusted in a municipal waste incinerator.</li> <li>Solidification of fly ash and landfilling of residues.</li> <li>Liquid digestate is spread on agricultural areas after hygienization.</li> </ul>	<ul> <li>Analogous to treatment path "AD+comp", but all CHP plants are located on-site for providing heat for drying.</li> <li>Solid digestate is not composted but pre-dried onsite to a DM of 40 wt%</li> <li>Transport and combustion of dried digestate in municipal waste incinerator.</li> <li>Solidification of fly ash and landfilling of residues.</li> </ul>	<ul> <li>Analogous to treatment path "AD", but the pasteurization step is excluded.</li> <li>Solid digestate is not spread but dried to a DM-content of 40 wt%</li> <li>Transportation to and combustion in a fluidized bed incinerator (mono-incineration).</li> <li>P is recovered from the ash.</li> <li>Landfilling of residues.</li> </ul>
" AD+G"	Anaer. Dig. + Gasification	N/A	<ul> <li>Analogous to treatment path "AD+I", but the dried, solid digestate is pelletized and gasified in a fixed bed gasifier (~800°C)</li> <li>Solidifying of fly ash and landfilling of residues</li> </ul>	<ul> <li>Analogous to treatment path "AD+comp", but solid digestate is pre-dried to reach a DM-content of &gt;85 wt%</li> <li>Dried digestate is pelletized and gasified in a fixed bed gasifier (~800°C)</li> <li>Solidifying of fly ash and landfilling of residues</li> </ul>	<ul> <li>Analogous to treatment path "AD+I", but digestate is dried to reach a DM-content of 70 wt%</li> <li>Gasification of the product in a fluidized bed gasifier (~800°C)</li> <li>P is recovered from the ash</li> <li>Landfilling of residues</li> </ul>

	Waste wood	Food waste	OFMSW	Sewage sludge
Anaerobic Digestion + HTC + Incineration	N/A	<ul> <li>Analogous to treatment path "AD", but digestate is separated into a solid and liquid fraction.</li> <li>Liquid digestate (DM=6%) is spread on agricultural areas after hygienization.</li> <li>Solid digestate is mixed to reach a biomass: water content of 0.15.</li> <li>Digestate undergoes HTC (200°C, 3h).</li> <li>Dewatering of suspension to ~60 wt% DM-content.</li> <li>Solid part is dried from 60 wt% to 90 wt% and is pelletized.</li> <li>HTC-char is sent to a centralized lignite power plant.</li> <li>Liquid part undergoes a cascade of nanofiltration and reverse osmosis.</li> <li>Retentate is either recirculated or sent to an incineration unit.</li> <li>Permeate is sent to a WWTP.</li> </ul>	<ul> <li>"AD+comp", but all CHP plants are located onsite and solid digestate is not composted.</li> <li>Solid digestate is mixed to reach a biomass:water content of 0.15.</li> <li>Digestate undergoes HTC (200°C, 3h).</li> <li>Dewatering of suspension to ~60 wt% DM-content</li> </ul>	<ul> <li>Analogous to treatment path "AD", but a pasteurization step is excluded.</li> <li>Dewatered sludge is mixed to reach a biomass: water content of 0.15.</li> <li>Digestate undergoes HTC (200°C, 3h).</li> <li>Dewatering of suspension to ~60 wt% DM-content</li> <li>Solid fraction is sent (without any further treatment) to a fluidized bed incinerator (mono-incineration)</li> <li>P is recovered from the ash.</li> <li>Landfilling of residues.</li> <li>Liquid fraction undergoes a cascade of nanofiltration and reverse osmosis.</li> <li>Retentate is either recirculated or sent to an incineration unit.</li> <li>Permeate is sent back to the WWTP</li> </ul>

		Waste wood	Food waste	OFMSW	Sewage sludge
" AD+HTC+G"	Anaerobic Digestion + HTC + Gasification	• N/A	Analogous to treatment path "AD+HTC+I", but pelletized char is gasified in a fixed bed gasifier (~950°C).  Syngas is co-combusted with biogas in an onsite CHP (250 kW) and a satellite plant (250 kW) for heat export to district heating system.  Solidification of fly ash and landfilling of residues.	"AD+HTC+I", but pelletized char is gasified in a fixed bed gasifier (~950°C).	<ul> <li>Analogous to treatment path "AD+HTC+I", but pelletized char is gasified in a fixed bed gasifier (~950°C).</li> <li>P is recovered from the ash.</li> <li>Solidification of fly ash and landfilling of residues.</li> </ul>
" HTC+G"	HTC + Gasification	<ul> <li>Analogous to treatment path "HTC+I", but pelletized char is gasified in a fixed bed gasifier (~950°C).</li> <li>Syngas utilization in satellite CHP plants (2x500 kW).</li> <li>Solidification of fly ash and landfilling of residues.</li> </ul>	Analogous to treatment path "HTC+I", but pelletized char is gasified in a fixed bed gasifier (~950°C).  Syngas utilization in an onsite CHP plant (300 kW).	<ul> <li>Analogous to treatment path "HTC+I", but pelletized char is gasified in a fixed bed gasifier (~950°C).</li> <li>Solidification of fly ash and landfilling of residues.</li> </ul>	N/A
" AD+P+I"	Anaer. Dig. + Pyrolysis + Incineration	N/A	N/A	N/A	<ul> <li>Analogous to treatment path "AD", but solid digestate is pre-dried to reach a DM-content of 22 wt%.</li> <li>Sewage sludge is pyrolyzed (500°C, 1 h).</li> <li>Pyrolysis-gas combustion in combustion chamber and utilization of heat for dryer.</li> <li>Char is transported to and combusted in a fluidized bed incinerator (mono-incineration).</li> <li>P is recovered from the ash.</li> <li>landfilling of residues.</li> </ul>

# Appendix I. Modelling of the incineration process

The incineration of organic waste or its secondary derivatives were largely based on simulation tools provided by DOKA (2017). The model describes the incineration process within a modern waste incinerator (grate-type) and calculates the energy production based on the elemental composition and the LHV. The LHV of liquid and solid fuels was calculated according to MICHEL (1938). Equation A. 1 states the calculation basis.

$$LHV_{s}\left[\frac{MJ}{kg}\right] = -9.8324\xi_{O} + 124.265\xi_{H} + 34.016\xi_{C} + 19.079\xi_{S} + 6.276\xi_{N}$$

$$-2.2\xi_{H2O} - 2.2 \cdot 9\xi_{H}$$
Equation A. 1

 $\xi$  = mass shares of the respective elements.

Parasitic electrical and thermal energy demands were accurately computed based on the waste type. As the model was based on grate incinerators, the parasitic electricity demands for fluidized bed incinerators was adjusted according to Neuwahl et al. (2019). For waste incineration (both grate and fluidized bed incinerators), a gross electric efficiency of ~16% and a gross thermal efficiency of ~60% with respect to the LHV, were set as boundary conditions. Steam parameters were herein 400°C and 40 bars and the overall combustion efficiency was ~75%. This represents typical values for waste incinerators that convert the combustion heat within a cogeneration unit, while featuring a single-stage condensation turbine (UBA 2018; FEHRENBACH ET AL. 2007).

For lignite power plants, gross electrical efficiencies of 42% and net electrical efficiencies of 38% with regards to the LHV of the substrate were implemented. This represents the conversion efficiency of a typical lignite power plant (UMWELTBUNDESAMT 2017). To achieve such high efficiencies, steam is expanded to ~0.025-0.035 bar and ~20-25°C (FLOSDORFF AND HILGARTH 2000). In this case, an exploitation of heat is not possible anymore.

For sewage sludge, P was recovered from the ash prior to its landfilling. P recovery according to the approaches of AshDec, Leachphos, EcoPhos, RecoPhos, PASCH and the Fertilizer Industry were used at equal shares (market mix). The energy demands were taken from AMANN ET AL. (2018).

# Appendix J. Modelling of the Anaerobic Digestion process

Subsequently, the modelling approaches for the processes, which were relevant in the context of anaerobic digestion are given.

#### a. Thickening

This process is only relevant for sewage sludge. A gravity thickener was implemented. The electricity consumption was ~100 kWh/t DM (DWA 2015; HONG ET AL. 2009; PETERS AND ROWLEY 2009; MILLS ET AL. 2014; RAMACHANDRAN ET AL. 2017).

#### b. Anaerobic digestion

Biogas yields and methane shares were based on empirical values. They are presented in the Electronic Annex for the considered substrates. To determine the composition of the resulting digestate, the Buswell-Müller (Buswell And Mueller 1952) equation was applied (Equation A. 2). The pathways of nitrogen and sulfur to ammonia and hydrogen sulphide can be neglected in this realm. Their shares in the biogas lie in the range of 0-4000 ppm (Sun et al. 2015).

$$C_a H_b O_c + \left(a - \frac{b}{4} - \frac{c}{2}\right) H_2 O \rightarrow \left(\frac{a}{2} + \frac{b}{8} - \frac{c}{4}\right) C H_4 + \left(\frac{a}{2} - \frac{b}{8} + \frac{c}{4}\right) C O_2$$
 Equation A. 2

C, H, O=carbon, hydrogen and oxygen of the fermentable biomass, a,b,c= molar shares

To solve Equation A. 2, the share of  $H_2O$  has to be known. REINHOLD (2006) suggests a value of 15-18% for agricultural biomass. A share of 16.5% was chosen for all calculations.

Anaerobic digesters and the respective infrastructure (e.g. mixers, dehydration units, conveyors, CHP plants) required heat and electricity during their operations. The parasitic electricity demand ranges between 2-10% of the produced electricity in the CHP plant (DACHS AND REHM 2006; BÖTTCHER 2013; Wellinger 2013). Regardless of the biogas plant size, an average value of 7.8% was determined. For digestion towers, operating on sewage sludge, an electricity demand of 1.6-2.3 kWh/m³ was used (PINNEKAMP ET AL. 2017).

The heat demands of anaerobic digestion plants was calculated individually based on the geometry of the plant and the process design. Heat was required for heating up the substrate plus water to the operational temperature, i.e. 37°. Further heat was needed to compensate heat losses due to radiation, conduction and convection. Heat gains through solar radiation were neglected. Biogas production resulted in an overall exothermic reaction, but the reaction enthalpy and thereof the heat production is negligible (PERRIGAULT ET AL. 2012). The calculation procedure to determine the heat demand is given in full detail in MAYER ET AL. (2020).

## c. CHP-plant

The combustion of biogas within CHP-plants within an internal combustion engine was adapted to an overall net efficiency of 90%. The net electric efficiency was based on WALLA AND SCHNEEBERGER (2008) and was 37% for a CHP-plant for a rated power of 500 kW<sub>el</sub>. The remaining, useful energy consisted of high temperature heat, which was extracted from the flue gas and low temperature heat, which resulted from the water and oil circuit to cool the engine. Following guidelines of ASUE (2014), the share of high temperature heat was 42%, while the share of low temperature heat was 58%.

#### d. Pasteurization

During the pasteurization of liquid streams heat was required to heat up the liquid to 71°C and to hold the temperature for 30 minutes. Equation A. 3 was utilized to determine the heat demand (KLAGES ET AL. 2009).

$$\dot{Q}_{past} = -\left(\dot{m}_{liquid} \cdot c_p \cdot \left(71^{\circ}\mathcal{C} - T_{digester} + 5^{\circ}\mathcal{C}\right)\right) \cdot 1.05 \cdot 0.3$$
 Equation A. 3

 $\dot{Q}_{past}$ =heat demand for pasteurization,  $\dot{m}_{liquid}$  = mass flow of liquid stream,  $c_p$ =heat capacity of liquid stream,  $T_{digester}$ =temperature in digester, i.e. 37°C, +5°C accounts for temperature losses between digester and pasteurization unit, 0.3 accounts for heat recovery (70% of heat can be recovered via heat exchangers), 1.05 accounts for heat losses in the pasteurization unit

#### e. Solid-liquid separation

It was assumed that the elemental composition was equal for the solid and liquid streams with regards to their dry mass. Electricity demands were not individually accounted for as they were already included in the sub-process anaerobic digestion.

# f. Dryer

A generic belt dryer was utilized for the drying of digestate. An average heat demand of 920 kWh/t  $H_2O_{evaporated}$  (BENNAMOUN ET AL. 2013) was implemented. The electricity demand was set to 60 kWh/t  $H_2O_{evaporated}$  (HEINDL 2016). Equation A. 4 applies.

$$\dot{Q}_{dryer,th/el} = \left(1 - \frac{DM_{in}}{DM_{target}}\right) \cdot \dot{m}_{digestate} \cdot \dot{q}_{th/el}$$
 Equation A. 4

 $\dot{Q}_{dryer,th/el}$ =thermal or electrical energy demand, DMin=dry matter content of inflowing stream, DMtarget= targeted dry matter content of the outflowing stream,  $\dot{m}_{digestate}$ =mass flow of digestate,  $\dot{q}_{th/el}$ =specific heat or electricity demand (i.e. 920 kWh<sub>th</sub> and 60 kWh<sub>el</sub>/t H<sub>2</sub>O)

## Appendix K. Modelling of the HTC process

A hydrothermal carbonization unit was previously modelled and the calculation steps were published in STOBERNACK ET AL. (2020). Implemented electricity and heat demands substrate specific and can be found in the Electronic Annex. This chapter summarizes the modelling steps towards determining the energy demands and mass distribution.

#### a. Hydrothermal Carbonization - energy demand

Components that require energy within the HTC process are the mixing tank (substrate + water), the grinder, the compressor pump and the HTC-reactor itself. An electricity demand of 20-38 kWh/t DM was implemented for the grinder and mixing unit (LUCIAN AND FIORI 2017). The compressor pump was calculated as a reversible work of a steady-state flow with an overall efficiency of 65%. Equation A. 5 can be used to determine the pump work. To simplify the calculation, it was assumed that the compression was isochoric.

$$W_{pump} = -\dot{m} \frac{1}{\eta_{pump}} \int \upsilon dp = -\dot{m} \frac{\upsilon}{\eta_{pump}} \cdot (p_{sat,T} - p_a)$$
 Equation A. 5

 $W_{pump}$ =work of the pump, m=mass flow of the suspension,  $\eta_{pump}$ =efficiency of the pump, i.e. 0.65,  $\nu$ =specific volume of suspension,  $p_{sat,T}$ =saturated steam pressure at 200 and 220 °C, i.e. 15.54 bar and 23.19 bar,  $p_a$ =ambient pressure, i.e.1 bar

The heat demand of the HTC-plant consisted of heating up the substrate and compensating heat losses. The reaction enthalpy of the HTC-process ranged between -1.1 to 0.6 MJ/kg DM (REBLING ET AL. 2015; FUNKE AND ZIEGLER 2011; JAN STEMANN; YAN ET AL. 2010) and may partially offset the heat demand. A share of the heat was recovered via heat exchangers. Equation A. 6 gives the total heat demand.

$$\dot{Q}_{heat} = -(T_{HTC} - T_a) \cdot \dot{m} \cdot c_p|_{T_a, p_a}^{T_{HTC}, p_{HTC}} - \dot{Q}_{loss} - \dot{Q}_{reaction} + \dot{Q}_{recover}$$
 Equation A. 6

 $T_{HTC}$ =temperature within the HTC reactor,  $T_a$ =ambient temperature,  $\dot{m}$ =mass flow of the suspension,  $\dot{Q}_{loss}$ = heat losses due to convection and radiation,  $\dot{Q}_{recover}$ = recovered heat from the exiting suspension,  $\dot{Q}_{reaction}$ =heat of reaction

Heat losses result from convection and radiation and are essentially depended on the geometry and the insulation material and thickness. For well insulated HTC-reactors these heat losses are negligible. The heat of reaction was assumed to be the difference between the heating value of the substrate and the products.

$$\dot{Q}_{reaction} = \sum \dot{m}_{product} \cdot LHV_{i,products} - \sum \dot{m}_{substrate} \cdot LHV_{substrate}$$
 Equation A. 7

 $\dot{Q}_{reaction}$ =heat of reaction,  $\dot{m}_x$ =mass flow of the substrate and respective product streams, LHV<sub>x</sub>= lower heating value of the substrate and the product streams.

Heat recovery was modelled according to BUTTMANN (2011). Indirect heat transfer was assumed. It is suggested that a suspension exiting the reactor at 200°C can be cooled down to 90°C and the respective heat can be recovered. For a reaction temperature of 220°C the suspension could be cooled down to 96°C (assuming the same geometry for the heat exchanger).

$$\dot{Q}_{recover} = \dot{m} \cdot c_{p,suspension} |_{t_{cool,p\,cool}}^{t_{reactor},p_{sat}} \cdot (t_{reactor} - t_{cool}) \cdot \eta_{HT}$$
 Equation A. 8

 $\dot{Q}_{recover}$ =recovered heat from the exiting suspension,  $\dot{m}$ =mass flow of suspension,  $c_p$ =integral heat capacity of the suspension,  $t_{reactor}$ =temperature in reactor (i.e. 200°C/220°C),  $t_{coof}$ =temperature after heat transfer (i.e. 90°C/96°C),  $\eta_{HT}$ =efficiency of heat transfer (i.e. 0.95)

The electricity demand for mixing the suspension within the HTC-plant was calculated for an anchor impeller. The rotational speed was set to 15 rounds per minute and the overall efficiency of the impeller was 80%. The correlation power number of the impeller was 3/Re^0.2, where Re is the Reynolds number (VDI 2013).

#### b. Hydrothermal Carbonization – mass distribution

The mass and elemental distribution during the HTC-process was based on empirical datasets. If applicable, parameter studies that varied operational parameters (retention time and temperature) were evaluated. Consequently, elemental and mass yields were extracted for the relevant operational parameters. Datasets were evaluated against the reaction intensity of the process. "The reaction intensity f (b) establishes a quantitative correlation between the reaction kinetic and the process temperature and retention time." (STOBERNACK ET AL. 2020). The equation to calculate the reaction intensity is given in Equation A. 9.

$$f(b) = 50 \cdot t^{0.2} \cdot e^{\left(-\frac{3500}{T}\right)}$$
 Equation A. 9

f(b)=reaction intensity [], t=retention time [s], T=reaction temperature [K]

Linear regression proved to be an apt approach to correlate elemental and mass yields against the reaction intensity. Figure A. 2 shows an exemplary evaluation for the HTC of sewage sludge. Based on the elemental yields in the char and the mass yields of the solid and gaseous phase (assumed as CO<sub>2</sub>), the elemental composition of the process water could be determined. The distribution of ash components was based on default values presented in REZA ET AL. (2013) and BERGE ET AL. (2015).

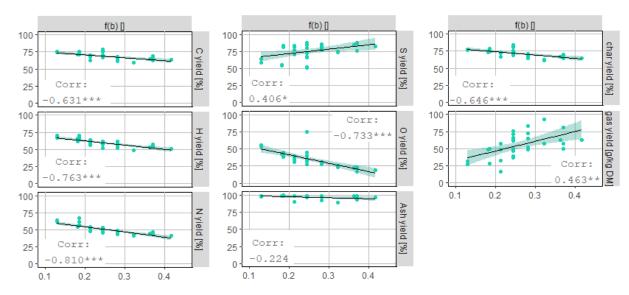


Figure A. 2: Elemental yields in char and mass yields during the HTC of sewage sludge in dependence of the reaction intensity f(b). Raw data based on BLÖHSE (2017).

For the HTC-process, a boundary condition was implemented that the H/C and O/C ratios were similar to those of lignite, if they were co-combusted in a lignite power plant. This was validated in a van-Krevelen diagram. Figure A. 3 gives the evaluation.

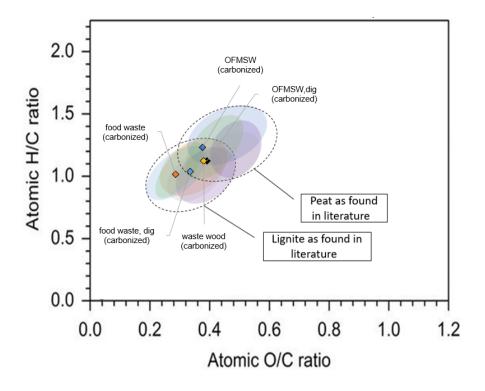


Figure A. 3: Depiction of the carbonized substrates in the van-Krevelen Diagram (Sources for lignite and peat window: Mendoza Martinez et al. (2021), Itoh et al. (2018), Knappe et al. (2019b), Pala et al. (2014))

#### c. Solid-liquid Separation

A solid-liquid separation of the suspension was modelled according to a decanter. The power demand was calculated according to LUCIAN AND FIORI (2017). The efficiency of the motor was assumed to be 85%. The diameter of the decanter was 0.43 m and the rotation speed was 5000 rpm (RECORDS AND SUTHERLAND 2001). The DM content of the solid stream after the separation step was assumed to be 60 wt.-%.

#### d. Dryer and pelletizer

The energy demand of the drying unit was calculated according to Equation A. 4. The char was dried to DM >90 wt.-%, if it was combusted in a lignite power plant and to DM>85 wt.-%, if it was gasified on-site. HTC was pelletized for further utilization. The electricity demand of the pelletizer was implemented at 96 kWh/t of char (WERNET ET AL. 2016).

#### e. Nanofiltration/Reverse osmosis

The process water exiting the HTC reactor is highly polluted and requires pre-treatment prior to a discharge to the sewer system. The share of organic pollutants is particularly high and thereof the COD is often used as indicator. A cascade of nanofiltration and reverse osmosis is proposed to treat the wastewater. Within the module the retentate and permeate was recirculated within certain boundary conditions, to reduce the amount of discharged water. Permeate which could not be recirculated was sent to a WWTP, while retentate was incinerated in a municipal waste incinerator (BILITEWSKI AND HÄRDTLE 2013).

Initially, the water treatment system was aligned to STARK AND MAAS (2012). The chemical oxygen demand (COD) value after the decanter was set to 83.1 g O2/kg process water, the

COD after both the filtration and the reverse osmosis were 150 g O2/kg process water. Separation efficiencies, i.e. ratio between retentate:permeate, were 47:53 for the nanofiltration module and 15:85 for the reverse osmosis module. The solid matter content of the OFMSW after the mixing tank was set to 15% (i.e. biomass:liquid=0.176). Exemplary, Figure A. 4 gives the outline for the treatment of 1 kg OFMSW (fresh matter).

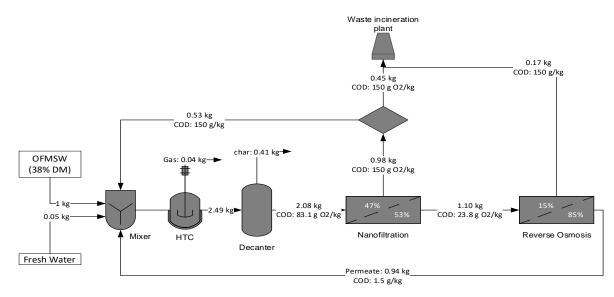


Figure A. 4: Process water treatment scheme. Values reference to the treatment of 1 kg of OFMSW (fresh matter) and the HTC operation parameters of 200°C and a retention time of 3 h.

The coal and gas yields were assumed not to be affected by the recirculation. The theoretical oxygen demand (ThOD) was used as proxy for the COD. Mass shares of organic matter and ash in the process water were assumed to not change throughout the system. Furthermore, it was assumed that the specific HTC-char and process gas yields were not affected by recirculation. The energy consumptions were estimated to be 2.1 kWh/m³ for the nanofiltration module (Bolle and Pinnekamp, 2011) and 7.1 kWh/m³ for the reverse osmosis module (Marshall, 2018).

The ThOD was calculated from the mass shares ( $\xi$ ) of C, H, O and N in the process water (PW). Equation A. 10 shows the calculation.

$$ThOD = COD = \left(\frac{\xi_{C,PW}}{12} + \frac{\xi_{H,PW}}{4} - \frac{\xi_{O,PW}}{8} - 0.75 \cdot \frac{\xi_{N,PW}}{14}\right) \cdot 32$$
 Equation A. 10

As described in the previous section, the ThOD of the process water after the decanter (ThOD<sub>PW</sub>) was predefined.

$$ThOD_{PW} = 83.1 \frac{g O_2}{kg PW}$$
 Equation A. 11

Also, the ThOD of the retentate was predefined.

$$ThOD_{retentate,recy} = ThOD_{retentate,NF} = ThOD_{retentate,RO} = 150 \ \frac{g \ O_2}{kg \ PW}$$
 Equation A. 12

The indices refer to the retentate from the nanofiltration (NF), reverse osmosis (RO) and the recirculated retentate (recy). The throughput (tp) of the substrate can be expressed as dry matter, according to Equation A. 13.

$$\dot{m}_{sub,DM} = DM \cdot \dot{m}_{tp}$$
 Equation A. 13

The remaining share of the throughput could be allocated to water.

$$\dot{m}_{sub.H2O} = (1-DM) \cdot \dot{m}_{tp}$$
 Equation A. 14

Within the mixer the substrate was diluted to a dry matter content of 15% or a biomass to liquid ratio (BL) of 0.176

$$BL = \frac{0.15}{1 - 0.15} = 0.176 \, kg/kg$$
 Equation A. 15

By implementing the gas and char yields (Y), the mass flows of gas and char could be calculated according to Equation A. 16 and Equation A. 17.

$$\dot{m}_{gas} = \dot{m}_{sub,DM} * Y_{gas}$$
 Equation A. 16

$$\dot{m}_{char,DM} = \dot{m}_{sub,DM} * Y_{solid}$$
 Equation A. 17

The dry matter content of the char after the decanter was 60% and thus the mass flow of the raw HTC-char is calculated according to Equation A. 18.

$$\dot{m}_{char,FM} = rac{\dot{m}_{char,DM}}{0.6}$$
 Equation A. 18

The share of dissolved matter in the process water without recirculation ( $DM_{PW}^*$ ) could be calculated in accordance to Equation A. 19.

$$DM_{PW}^* = (\dot{m}_{OFMSW,DM} - \dot{m}_{char,DM} - \dot{m}_{aas})/\dot{m}_{PW,out}$$
 Equation A. 19

By closing the mass balance, the amount of process water exiting the decanter ( $\dot{m}_{PW,out}$ ) could be calculated.

$$\dot{m}_{PW,out} = \dot{m}_{sub,DM} + \dot{m}_{sub,H20} + \dot{m}_{H20,fresh} + \dot{m}_{permeate,recy} + \dot{m}_{retentate,recy} - \dot{m}_{char,FM} - \dot{m}_{gas}$$
 Equation A. 20

where  $\dot{m}_{H20,fresh}$  is the fresh water, which is added to the mixer.  $\dot{m}_{permeate,recy}$  and  $\dot{m}_{retentate,recy}$  are the shares of recirculated permeate and retentate.

The mass flow of retentate ( $\dot{m}_{retentate,recy}$ ) that can be recirculated was limited by the COD exiting the decanter (i.e. 83.1 g O<sub>2</sub>/kg PW). The recirculated amount could be calculated according to Equation A. 21.

$$\dot{m}_{retentate,recy} = ((ThOD_{PW} - ThOD \cdot DM_{PW}^*) \cdot \left(\frac{\dot{m}_{OFMSW,DM}}{BL}\right) ... \\ - (\dot{m}_{permate,recy} \cdot ThOD_{permeate,recy}))/ThOD_{retentate,recy}$$
 Equation A. 21

The amount of fresh water that had to be added to the system was dependent on the amount of water which was recirculated and the biomass:liquid ratio. Equation A. 22 details the calculation.

$$\dot{m}_{H_2O,fresh} = \dot{m}_{sub,DM}/BL - \dot{m}_{permeate,recy} - \dot{m}_{retentate,recy} - \dot{m}_{sub,H2O}$$
 Equation A. 22

The mass flow distributions between retentate and permeate during the nanofiltration and reverse osmosis are dependent on the separation efficiencies. Equation A. 23-Equation A. 26 implement the distribution factors. It was assumed that the permeate exiting the reverse osmosis module was completely recirculated. If a full recirculation of the permeate was not possible, the remainder was sent to a WWTP.

$$\dot{m}_{permeate,recy} = \dot{m}_{permeate,RO} = \dot{m}_{PW,out} \cdot 0.53 \cdot 0.85$$
 Equation A. 23 
$$\dot{m}_{permeate,NF} = \dot{m}_{PW,out} \cdot 0.53$$
 Equation A. 24 
$$\dot{m}_{retentate,NF} = \dot{m}_{PW,out} \cdot 0.47$$
 Equation A. 25 
$$\dot{m}_{retentate,RO} = \dot{m}_{PW,out} \cdot 0.53 \cdot 0.15$$
 Equation A. 26

The calculation of the ThOD of the permeates after the nanofiltration and reverse osmosis was calculated by distributing the ThOD of the inflow to the retentate and permeate.

$$ThOD_{permeate,NF} = (\dot{m}_{PW,out} \cdot ThOD_{PW} - \dot{m}_{retentate,NF})$$

$$= (\dot{m}_{PW,out} \cdot ThOD_{PW} - \dot{m}_{retentate,NF})$$

$$\cdot ThOD_{retentate,NF}) / \dot{m}_{permeate,NF}$$

$$ThOD_{permeate,RO} = (ThOD_{permeate,NF} \cdot \dot{m}_{permeate,NF} - ThOD_{retentate,RO})$$

$$\cdot \dot{m}_{retentate,RO}) / \dot{m}_{permeate,RO}$$
Equation A. 28

The share of retentate sent to a municipal waste incinerator was the retentate from both the nanofiltration and reverse osmosis minus the recirculated retentate (Equation A. 29)

$$\dot{m}_{inc} = \dot{m}_{retentate,NF} + \dot{m}_{retentate,RO} - \dot{m}_{retentate,recv}$$
 Equation A. 29

After this initial setup, the treatment system was adjusted to the following boundary conditions:

- The dissolved matter content after the nanofiltration and the reverse osmosis was set to 15 wt.-%.
- The COD after the reverse osmosis was set to 1.5 gO<sub>2</sub>/L.

This became necessary to account for varying ash contents.

# Appendix L. Modelling of the pyrolysis process

#### a. Dryer

If necessary, substrate was pre-dried to enhance the overall plant efficiency. The energy demand was calculated according to Equation A. 4.

## b. Pyrolysis - energy demand

The energy demand was composed of an electric energy demand for the pyrolysis reactor and the respective infrastructure (conveyor, shredder, flue gas treatment) and a heat demand for the rotary kiln. The electricity demand was taken from a former pyrolysis plant in Burgau, Germany and was 0.18 kWh/kg treated waste (THOMÉ-KOZMIENSKY AND BECKMANN 2014). The electricity demand is therefore similar to those of incineration plants (NEUWAHL ET AL. 2019) and appears plausible. Pyrolysis was modelled for a retention time of 60 minutes and a temperature within the rotary kiln of 500°C. Thermal energy was required for heating up the substrate, heating up and evaporating water and to counteract heat losses. Furthermore, the reaction enthalpy was accounted for. It was calculated according to Equation A. 7. The total heat demand was calculated according to Equation A. 30.

$$\begin{split} \dot{Q}_{heat} &= - \left( T_{pyro} - T_a \right) \cdot \dot{m}_{dry} \cdot c_p \big|_{T_a}^{T_{pyro}} - \dot{Q}_{loss} - \dot{Q}_{reaction} - c_{p,H2O} \big|_{T_a}^{100^{\circ}C} \\ & \cdot \dot{m}_{H_2O} \cdot (373.15K - T_a) - \dot{m}_{H_2O} \cdot \Delta_{\rm E} H^{\circ} - c_{p,H2O} \big|_{100^{\circ}C}^{T_{pyro}} \cdot \dot{m}_{H_2O} \\ & \cdot \left( T_{pyro} - 373.15K \right) \end{split} \label{eq:Qheat}$$
 Equation A. 30

 $T_{pyro}$ =temperature within the pyrolysis chamber,  $T_a$ =ambient temperature,  $\dot{m}_{dry}$ =mass flow of the dry substrate,  $c_p$ =integral heat capacity of the substrate, liquid water and gaseous water,  $\dot{Q}_{loss}$ = heat losses due to convection and radiation, i.e. assumed to be 5% of the total heat production,  $\dot{m}_{H20}$ =mass flow of water in the substrate,  $\Delta_E H^\circ$ =standard enthalpy of evaporation of water,  $\dot{Q}_{reaction}$ =heat of reaction

#### c. Pyrolysis - mass distribution

During pyrolysis, a gaseous phase emerged from the pyrolysis process, consisting of non-condensable gases, an aqueous oil phase and a heavy oil phase. As all components were in a gaseous state at the respective operational parameters, it is referred to as pyrolysis gas. The char was discharged from the rotary kiln. The distribution of elements and the char, oil and gas yields were taken from literature data. Despite a partly weak data basis on the elemental distribution the different components, it was assumed that only minor errors were introduced in the model, as the gas and oil phase were utilized together. The distribution of elements for the respective substrates is given in section B.6 of the Electronic Annex.

#### d. Utilization of pyrolysis gas

Pyrolysis gas (incl. oil) was combusted in a combustion chamber. The adiabatic combustion temperature was set to 1250°C, while  $\lambda$ =1.3-1.5. A share of the heat stream was drawn to heat the pyrolysis reactor and the cooled down stream was finally mixed again with the hot flue gas stream (an outline is given in MAYER ET AL. (2021)). Depending on the substrate, two options were given:

- 1. Option: the heat was only utilized to satisfy the heat demand of the dryer. Therefore, the heat of the flue gas was channeled through a heat exchanger.
- 2. Option: the heat was utilized to drive a condensation turbine in the realm of a Rankine Cycle.

Subsequently, the calculation steps for the utilization of pyrolysis gas and oil are described. Pyrolysis gas and oil were combusted together. The combustion process was largely based on HELLFRITSCH AND KOPPE (2007). The flue gas composition and the minimal oxygen demand  $(O_{2,min})$  were calculated according to Table A. 3.

Table A. 3: Calculation of the flue gas composition and the minimal oxygen demand

flue gas	CO2	H2O	N2	HCI	HF	SO2	O <sub>2</sub> ,min
Fuel							
$\dot{n}_{ash}$							
$\dot{n}_C$	$\dot{n}_C$						$\dot{n}_C$
$\dot{n}_H$		$0.5 \times \dot{n}_H$					$\dot{n}_H/4$
$\dot{n}_O$							$\dot{n}_{O}/2$
$\dot{n}_N$			$0.5 \times \dot{n}_N$				
$\dot{n}_S$						$\dot{n}_S$	$\dot{n}_S$
$\dot{n}_{Cl}$		-0.028x2x $\dot{n}_{Cl}$		$\dot{n}_{Cl}$			
$\dot{n}_F$		-0.053x2x $\dot{n}_F$			$\dot{n}_F$		
$\dot{n}_{H_2O}$		$\dot{n}_{H_2O}$					
$\dot{n}_{CH_4}$	$\dot{n}_{CH_4}$	2 x $\dot{n}_{CH_4}$					2 x $\dot{n}_{CH_4}$
$\dot{n}_{CO}$	$\dot{n}_{CO}$						$\dot{n}_{CO}/2$
$\dot{n}_{H_2}$		$\dot{n}_{H_2}$					$\dot{n}_{H_2}/2$
$\dot{n}_{C_2H_4}$	$2 \times \dot{n}_{C_2H_4}$	$2 \times \dot{n}_{C_2H_4}$					$\dot{n}_{C_2H_4} \times 3$
$\dot{n}_{C_2H_6}$	$2 \times \dot{n}_{C_2H_6}$	$3 \times \dot{n}_{C_2H_6}$					$\dot{n}_{C_2H_6} \times 3.5$
$\dot{n}_{N_2}$			$\dot{n}_{N_2}$				
SUM	$\Sigma = \dot{n}_{CO_2}$	$\sum \dot{n}_{H_2O}$	$\Sigma = \dot{n}_{N_2}$	$\sum = \dot{n}_{HCl}$	$\Sigma = \dot{n}_{HF}$	$\sum = \dot{n}_{SO_2}$	$\sum = \dot{n}_{O_{2,min}}$

Flue gas components as given in Table A. 3 were transferred into mass flows by multiplying them with their respective molar mass. The flue gas for an air-fuel-ratio  $\lambda$ =1, which resulted from the pyrolyzed substrate was calculated according to Equation A. 31.

$$\dot{m}_{c,wet} = \dot{n}_{CO_2} \cdot M_{CO_2} + \dot{n}_{H_2O} \cdot M_{H_2O} + \dots + \dot{n}_{O_2,min} \cdot M_{O_2}$$
 Equation A. 31

The amount of dry air which was needed for the combustion was calculated in dependence of the air-fuel ratio ( $\lambda$ ) and the minimal oxygen demand. An oxygen share of 21% in the ambient air was assumed.

$$\dot{n}_{air,dry} = \frac{\lambda \cdot \dot{n}_{O_2,min}}{0.21}$$
 Equation A. 32

$$\dot{m}_{air,dry} = \dot{n}_{air,dry} \cdot (0.79 \cdot 28.013 + 0.21 \cdot 32.000)$$
 Equation A. 33

The share of water in the air (x) was calculated according to Equation A. 34.

$$x \left[ \frac{kg}{kg} \right] = \frac{\varphi_{air} \cdot p_s}{R_s \cdot T}$$
 Equation A. 34

$$\dot{m}_{air,wet} = \dot{m}_{air,wet} \cdot (1+x)$$
 Equation A. 35

with  $m_{H2O}$  being the mass of water in the flue gas,  $\phi_{air}$  being the humidity of air,  $p_S$  being the density of saturated water pressure,  $R_S$  being the gas constant for water vapor, T being the air temperature [K] and FG<sub>wet</sub> being the amount of flue gas.

$$\dot{m}_{FG,wet} = \dot{m}_{c,wet} + x \cdot \dot{m}_{air,wet} + 0.79 \cdot \dot{n}_{air,dry} \cdot 28.013 + 0.21 \cdot \dot{n}_{air,dry}$$
 Equation A. 36 
$$\cdot 32.00 - \dot{n}_{O_2,min} \cdot M_{O_2}$$

The air-fuel ratio was adjusted to reach the desired combustion temperature, i.e. 1250 °C (adiabatic combustion temperature). A share of the flue gas was recirculated (subscript rezy) to control the temperature in the combustion chamber. The adiabatic combustion temperature was calculated according to Equation A. 37-Equation A. 41.

$$t_{ad} = 25 + \frac{H_1 + H_2 + H_3}{H_4} = t_{cc} = 1250$$
°C Equation A. 37

The term H₁ refers to the LHV of the oil and gas multiplied by their mass flows.

$$H_1 = (LHV_{gas} \cdot \dot{m}_{gas} + LHV_{oil} \cdot \dot{m}_{oil})$$
 Equation A. 38

 $H_2$  refers to the enthalpy due to the sensible heat of the gas, dry pyrolysis oil and the water in the oil. At the respective temperature ( $t_{gas,pyro}$  – temperature of the pyrolysis gas/oil when exiting the pyrolysis reactor and  $t_{gas,rezy}$  – temperature of the recirculated flue gas) all components are in a gaseous state.

$$H_2 = \dot{m}_{gas} \cdot c_{p,gas} \Big|_{25}^{t_{gas,pyro}} \cdot (t_{gas,pyro} - 25) + \cdots$$
 Equation A. 39

$$\begin{split} \dot{m}_{oil,H_2O} \cdot c_{p,H2O}|_{25}^{t_{gas,pyro}} \cdot \left(t_{gas,pyro} - 25\right) + \cdots \\ \dot{m}_{oil,dry} \cdot c_{p,oil,dry}|_{25}^{t_{gas,pyro}} \cdot \left(t_{gas,pyro} - 25\right) + \cdots \\ \dot{m}_{FG,rezy} \cdot c_{p,FG,wet}|_{25}^{t_{gas,rezy}} \cdot \left(t_{gas,rezy} - 25\right) \end{split}$$

The term H₃ refers to the enthalpy of the combustion air. No air preheating is assumed and the air temperature is thus the ambient air.

$$H_3 = \dot{m}_{air,dry} \cdot (c_{p,air,dry}|_{25}^{t_{air}} + x \cdot c_{p,H_2,0}|_{25}^{t_{air}}) \cdot (t_{air} - 25)$$
 Equation A. 40

H<sub>4</sub> describes the enthalpy of the flue gas at the adiabatic combustion temperature.

$$H_4 = \dot{m}_{FG,wet} \cdot c_{p,FG,wet}|_{25}^{t_{ad}} + \dot{m}_{FG,rezy} \cdot c_{p,FG,wet}|_{25}^{t_{ad}}$$
 Equation A. 41

The heat capacity of the flue gas was calculated according to Equation A. 42

$$\begin{aligned} c_p|_0^t &= 0.9647 + 0.7674 \cdot \xi_{\text{H}_2O} + 0.0248 \cdot \xi_{CO_2} + \cdots \\ & \left(9.3346 \cdot 10^{-5} + 1.7462 \cdot 10^{-4} \cdot \xi_{H_2O}^{0.8706}\right) \cdot T^{1.0189} \end{aligned}$$
 Equation A. 42

with T being the flue gas temperature in K and  $\xi$  being the mass shares of the elements in the flue gas. An iterative approach was necessary to calculate the heat capacity and the air-fuel ratio  $\lambda$ .

The reference temperature for the adiabatic temperature is 25°C. Values for the capacity are referenced for 0°C. The following adjustment is applicable.

$$c_p|_{25}^t = \frac{(t - 0^{\circ}C) \cdot c_p|_0^t - (25^{\circ}C - 0^{\circ}C) \cdot c_p|_0^{25}}{t - 25^{\circ}C}$$
 Equation A. 43

The total heat production could be calculated according to Equation A. 44

$$Q_{prod} = \left(\dot{m}_{FG,wet} \cdot c_{p,FG,wet}|_{25}^{tad} + \dot{m}_{FG,rezy} \cdot c_{p,FG,wet}|_{25}^{tad}\right) \cdot \dots \\ \cdot (t_{ad} - t_{ambient}) + \Delta_R H$$
 Equation A. 44

Heat capacity values of pure substances were sourced from ASTHER (2019). The heat capacity value of the inhomogeneous group of pyrolysis oil (without water) was assumed to be 1.65 kJ/kg/K (RIEDL 2007). Due to scarcity of data a constant value was taken.

For the heat utilization, two options were considered.

#### Option 1: Heat transfer via heat exchanger to dryer

The sensible heat of the flue gas was transferred to a heat exchanger and the temperature after the heat exchanger was assumed to be  $t_{FG}=200^{\circ}$ . By means of Equation A. 45 the energy losses due to sensible heat could be estimated.

$$\xi_{RG} = \frac{\dot{m}_{FG,wet} \cdot c_{p,FG,wet}|_{25}^{t_{FG}} \cdot (t_{FG} - 25^{\circ}C)}{H_{u} + \dot{m}_{air,drv} \cdot (c_{p,air,drv}|_{25}^{t_{air}} + x \cdot c_{p,H_{2}O}|_{25}^{t_{air}}) \cdot (t_{air} - 25^{\circ}C)}$$
 Equation A. 45

Further losses ( $\xi_{other}$ ) were in accordance to HELLFRITSCH AND KOPPE (2007) estimated at ~3% (accounting for radiation, convection etc.).

The parasitic heat demand had to be subtracted from the utilizable heat. It consisted of heating up and evaporating the water in the substrate (Equation A. 46), heating up the substrate itself (Equation A. 48), and heat losses in the rotary kiln (Equation A. 48). Data on heat losses were not given and were assumed to be 5% of the total produced heat.

$$\dot{Q}_{H2O} = \dot{m}_{substrate} \cdot (1 - DM) \cdot (H_{H_2O}^{t_{pyro}} - H_{H_2O}^{25^{\circ}C})$$
 Equation A. 46 
$$\dot{Q}_{substrate} = c_{p,substrate} \cdot (\dot{m}_{substrate} \cdot DM) \cdot (t_{pyro} - 25^{\circ}C)$$
 Equation A. 47 
$$\dot{Q}_{losses} = 0.05 \cdot (\dot{Q}_{prod})$$
 Equation A. 48

The heat demand was partially offset by the reaction enthalpy ( $\Delta_R H$ ) and the total heat output can be calculated according to Equation A. 49.

$$\dot{Q}_{tot} = \{Q_{prod} - (\dot{Q}_{H2O} + \dot{Q}_{substrate} + \dot{Q}_{losses})\} \cdot (1 - \xi_{RG} - \xi_{other})$$
 Equation A. 49

#### Option 2: Heat utilization to drive a steam turbine

The electricity output could be determined by means of a Rankine-Cycle. Figure A. 5 depicts the specific steam cycle in a T,s-diagram.

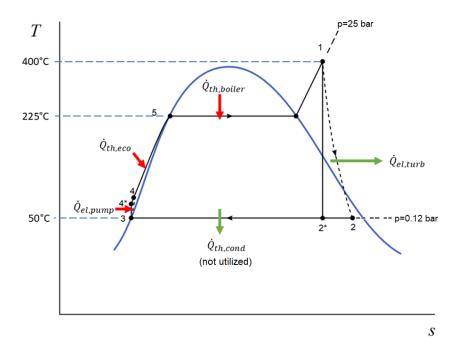


Figure A. 5: Rankine Cycle in T,s-Diagram. Parameter settings according to condensation turbine in pyrolysis plant (not to be scaled).

The efficiency of the cycle was calculated according to Equation A. 50.

$$\eta_{CR} = \frac{h_1 - h_2 - (h_4 - h_3)}{h_1 - h_4}$$
 Equation A. 50

where h is the specific enthalpy at the points 1-5 as outlined in Figure A. 5. The values provided in Table A. 4 were applied.

Table A. 4: enthalpies of points h<sub>1</sub> to h<sub>5</sub>

	T [°C]	p [bar]	state	spec. enthalpy [kJ/kg]
h <sub>1</sub>	400	25	superheated steam	3241
h <sub>2</sub>	50	0.12	saturated steam, x=0.9	2351
h <sub>3</sub>	50	0.12	water	209
h <sub>4</sub>	50	25	water	212
h <sub>5</sub>	225	25	water	961

The points  $h_2$  and  $h_5$  were calculated from the turbine efficiency ( $\eta_{\text{turbine}}$ =0.9) and pump efficiency ( $\eta_{\text{comp}}$ =0.89). Equation A. 51 and Equation A. 52 detail the calculation.

$$h_2 = (h_2^* - h_1) \cdot \eta_{turbine} + h_1$$
 Equation A. 51

$$h_4 = (h_4^* - h_3)/\eta_{comp} + h_3$$
 Equation A. 52

The total electricity production was determined by Equation A. 53.

$$\dot{Q}_{el,turb} = \eta_{CR} \cdot \dot{Q}_{tot} \cdot \eta_{gen}$$
 Equation A. 53

where  $\eta_{gen}$  is the efficiency of the generator, i.e.0.97.  $\dot{Q}_{tot}$  was the total heat production and may be taken from Equation A. 49. The gross condensation heat could be calculated from Equation A. 54, but the temperature level was not considered adequate for a heat export to a district heating system.

$$\dot{Q}_{th,cond} = rac{h_2 - h_3}{h_1 - h_4} \cdot \dot{Q}_{tot}$$
 Equation A. 54

#### Appendix M.Modelling of the gasification process

#### a. Dryer

Substrate was pre-dried to <15 wt.-% (fixed bed gasifier), or <30% (fluidized bed gasifier). Waste wood was not pre-dried. The energy demand for drying was calculated according to Equation A. 4.

#### b. Gasification

For bulky or compacted material, a downdraft fixed gasifier was proposed. Otherwise, a fluidized bed gasifier was used. The composition of the product-gas was based upon equilibrium modelling. Therefore, the temperature levels had to be defined initially. A gasification temperature of 800°C was assumed for sewage sludge (typical temperature for stationary fluidized bed gasifiers), and for all other substrates it was 950°C (given the problematic issue of ash

melting, the temperature was reduced for substrates with an unknown performance in a gasification unit). The product gas exiting the reactor was 700°C and was cooled down within a heat exchanger, whereby the inflowing air (gasification agent) was heated up to 300°. Figure A. 6 states a schematic sketch for both gasifier types.

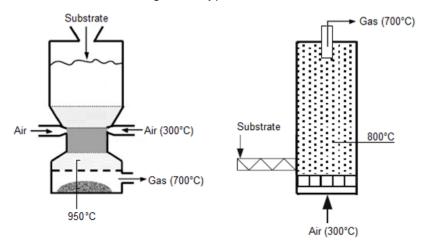


Figure A. 6: Schematic illustration of a fixed bed gasifier (left) and a fluidized bed gasifier (right). Artwork based on Kaltschmitt et al. (2016).

Input data for the gasification model were the elemental composition of the substrate and the gasification temperature. The substrate was corrected for the shares of tar in the product gas, i.e. 0.5 g/Nm³ for fixed bed gasifiers and 12 g/m³ for fluidized bed gasifiers (KALTSCHMITT ET AL. 2016). An average tar composition (CH $_{0.85}$ O $_{0.17}$  (BURAGOHAIN ET AL. 2013)) was assumed. Overall losses (heat losses, organic material, which was not gasified etc.) were assumed to be 10% of the average lower heating value of the substrate. To determine the heat losses due to ash discharge, the heat capacity of SiO $_2$  was used as a proxy for ash. The temperature of the ash was assumed to equal the gasification temperature. The heat capacity of C $_5$ H $_8$  was used as a proxy for tar. Equation A. 55 in conjunction with Equation A. 56 were the boundary conditions within this framework.

 $\dot{q}_{ash}$ = heat loss through ash (sensible heat),  $\dot{m}_x$ = mass flow of ash, tar, gas, air or substrate,  $\dot{Q}_{loss}$ =heat losses through convection and radiation, LHV<sub>x</sub>=lower heating value of tar or substrate,  $\dot{q}_{tar}$ =heat loss through tar (sensible heat),  $\dot{q}_{air}$ =sensible heat of inflowing air.

To close the energy balance, the gasification-air-fuel-ratio ( $\lambda$ ) had to be iterated to close the energy balance. A set of eight equations, based on the conservation of mass and the equilibrium conditions were implemented, to determine the shares of H<sub>2</sub>, H<sub>2</sub>O, CH<sub>4</sub>, CO<sub>2</sub>, CO, C<sub>2</sub>H<sub>4</sub> and C<sub>2</sub>H<sub>6</sub> and the specific product gas quantity ( $\dot{n}_{gas}$ ) during gasification.

$$\dot{n}_{gas} = \frac{\dot{n}_{H(substrate)}}{2 \cdot x_{H_2O} + x_{H_2} + 4 \cdot x_{CH_4} + 4 \cdot x_{C_2H_4} + 6 \cdot x_{C_2H_6}}$$
 Equation A. 57

$$x_{H_2O} = \frac{\dot{n}_{O(substrate)}}{\dot{n}_{gas}} - x_{CO} - 2 \cdot x_{CO_2}$$
 Equation A. 58 
$$x_{CO} = \frac{\dot{n}_{C(substrate)}}{\dot{n}_{gas}} - x_{CO_2} - x_{CH_4} - 2 \cdot x_{C_2H_4} - 2 \cdot x_{C_2H_6}$$
 Equation A. 59 
$$x_{CO} \cdot x_{H_2O} \cdot K_1$$

$$x_{CO_2} = \frac{x_{CO} \cdot x_{H_2O} \cdot K_1}{x_{H_2}}$$
 Equation A. 60

$$x_{CH_4} = 2 \cdot x_{H_2} \cdot \dot{n}_{C(substrate)} \cdot K_2$$
 Equation A. 61

$$x_{C_2H_4} = \frac{2 \cdot x_{CO_2} \cdot 2 \cdot x_{H_2O} \cdot \dot{n}_{gas}}{K_3 \cdot 6 \cdot \dot{n}_{O(substrate)}}$$
 Equation A. 62

$$x_{C_2H_6} = \frac{\left(2 \cdot x_{CO} \cdot \dot{n}_{gas}\right) \cdot \left(3 \cdot x_{H_2O}\right)}{5 \cdot \dot{n}_{O(substrate)} \cdot K_4}$$
 Equation A. 63

$$x_{H_2} = 1 - x_{H_2O} - x_{CO} - x_{CO_2} - x_{CH_4} - x_{C_2H_4} - x_{C_2H_6}$$
 Equation A. 64

 $\dot{n}_{C(substrate)}$ ,  $\dot{n}_{H(substrate)}$  and  $\dot{n}_{O(substrate)}$  were the molar masses in the substrate (corrected for the tar content). x was the molar shares of the respective elements. K<sub>1</sub>, K<sub>2</sub>, K<sub>3</sub> and K<sub>4</sub> are reaction specific equilibrium constants.

$$K_1 = \frac{x_{CO_2} \cdot x_{H_2}}{x_{CO} \cdot x_{H_2O}} = e^{\frac{\Delta r G_1^0}{R \cdot T}}$$
 Equation A. 65

$$K_2 = \frac{x_{CH_4} \cdot \dot{n}_{gas}}{2 \cdot x_{H_2} \cdot \dot{n}_{gas} \cdot \dot{n}_{C(substrate)}} = e^{\frac{\Delta r G_2^0}{R \cdot T}}$$
 Equation A. 66

$$K_3 = \frac{2 \cdot x_{CO_2} \cdot 2 \cdot x_{H_2O} \cdot \dot{n}_{gas}^2}{6 \cdot \dot{n}_{O(substrate)} \cdot x_{C_2H_6} \cdot \dot{n}_{gas}} = e^{\frac{\Delta r G_3^0}{R \cdot T}}$$
 Equation A. 67

$$K_4 = \frac{2 \cdot x_{CO} \cdot \dot{n}_{gas}^2 \cdot 3 \cdot x_{H_2O}}{5 \cdot \dot{n}_{O(substrate)} \cdot x_{C_2H_c} \cdot \dot{n}_{gas}} = e^{\frac{\Delta r G_4^0}{R \cdot T}}$$
 Equation A. 68

with R being the ideal gas constant, T being the temperature and  $\Delta r G^0$  being the change in the Gibbs enthalpy. Table A. 5 provides the equilibrium constants for the given equations and for the considered temperatures at atmospheric pressure.

Table A. 5: equilibrium constants for the given reactions (at atmospheric pressure) (AsTHER 2019)

Temperature [°C]	<b>K</b> 1	K2	K3	K4
800	1.02	0.045	1.11E+63	6.88E+53
950	0.67	0.012	1.34E+55	4.51E+48

The cold gas efficiency ( $\eta_{coldgas}$ ) was calculated according to Equation A. 69.

$$\eta_{coldgas} = \frac{LHV_{gas} \cdot \dot{m}_{gas}}{LHV_{substrate} \cdot \dot{m}_{substrate}}$$
 Equation A. 69

#### c. CHP-plant

The syngas was combusted within a CHP-plant. The same efficiencies as stated for the combustion of biogas (Appendix J) were applied for the combustion of syngas. As outlined in Appendix E, the initially lower heating value of syngas in comparison to biogas does not affect the combustion efficiency.

# Appendix N. Short summary on legal regulations in the realm of waste-to-energy

While the Circular Economy Law (KrWG) provides general guidelines for an environmentally friendly waste treatment, explicit regulations are defined in further ordinances and must be followed with regards to emissions resulting prior, during and past the treatment of waste in WtE plants. This section states the main regulations (without any claim to completeness) that have to be complied with, while operating large-scale WtE plants. Legal regulations for the authorization process are not covered.

Admissible environmental impacts, resulting from waste treatment plants are incorporated in the Federal Immission Control Act (BImSchG). According to §4(1) BImSchG and appendix 1, number 8 of the 4. Federal Immission Control Ordinance (BlmSchV), large-scale waste treatment plants and plants that allow for a co-combustion of hazardous and non-hazardous waste streams are subject to authorization and thus emission measurements have to be conducted regularly (§26-29 BlmSchG). For thermal treatment plants, the limit values are defined in the 17. BlmSchV and for biological waste treatment plants in the 30. BlmSchV. Within the 44. BImSchV limit values for the combustion of secondary gaseous fuels resulting from biological treatment process (including biogas and sewage gas) are found. The treatment by means of HTC (excluding the energetic utilization of HTC-char) is not yet explicitly regulated. According to appendix 1, number 8.8 of the 4. BlmSchV they require authorization. Currently, the produced chars are to be considered as waste and must thereof be treated in waste treatment plants according to the 17. BImSchV. With regards to waste wood of the classes AIII and AIV it should be noted that this waste stream is considered of hazardous nature. In principle, an energetic treatment in mono- and co-treatment plants with a high energy exploitation rate according to the R1-criterion is desirable according to the Waste Wood Ordinance (AltholzV). However, a more stringent regulation is given according to the §3 (3) of the 17. BImSchV for hazardous waste.

Table A. 6 summarizes the relevant extracts of the BImSchV in the realm of this work with regards to limit values. For all processes involved in the waste treatment that are not explicitly regulated in any BImSchV, limit values stated in the *Technical Instructions on Air Quality* (TALuft) must be maintained.

Table A. 6: Summary on the legislation for the operation of the considered WtE-plants (focus on limit values)

	17. BlmSchV	30. BlmSchV	44. BlmSchV
Application	incineration, gasification, pyrolysis (incl. combustion of secondary gas + char), co-combustion	anaerobic di- gestion, com- posting	combustion of biogas/sewage gas
threshold applica- tion (non-hazardous waste)	-	>10 t/day	>1 MW & <50 MW (rated thermal input)
Definition of limit values for emissions	§8 in conjunction with Appendix 1 & §10 for waste incinerators, pyrolysis plants and gasification plants;	<b>§</b> 6	§9, §16 (for combustion engines <10 MW rated thermal input)
	§9 in conjunction with Appendix 3 for co- combustion		

For by-products of waste treatment plants, further regulations exist. Waste waters from waste treatment plants are usually to be pre-treated, to meet the regulations given in *Waste Water Regulation* (AbwV). Appendix 23, 27 and 33 of the AbwV contain the relevant limit values that treated water from waste treatment plants has to comply with. According to § 12(1) of the 17. BlmSchV and §5(1) of the BlmSchG, solid residues from the thermal treatment, including ash and slag, must be avoided, recycled or disposed of. It must be ensured that slag and grate ash contain <3 wt.-% C and are characterized by an ignition loss <5 wt.-% (§5 of the 17. BlmSchV). Disposal of residues is comprehensively regulated in the *Landfill Ordinance* (DepV).

During the operation of anaerobic digestion plants, digestate is produced. Digestate may be spread on agricultural areas and limit values stated in Appendix 2 of the *Fertilizer Ordinance* (DüMV) have to be maintained. For the spreading of sewage sludge additional thresholds apply. They are given in Appendix 1 of the *Sewage Sludge Ordinance* (AbfKlärV). No more than 5 t (measured as DM) of sewage sludge may be spread on one hectare within 3 years. For biowaste acceptable contamination levels of treated OFMSW are regulated in §4(3) of the *Biowaste Ordinance* (BioAbfV). The spreading of organic fertilizer from OFMSW is limited to 20 t (measured as DM) per hectare within 3 years. On the account of fertilizer application, also §6 of the *Fertilizing Ordinance* (DüV) must be maintained, which states that less than 170 kg/ha of nitrogen must be applied via organic sources.

#### **Appendix O. Pedigree Matrix**

If the data quality of flows had to be assessed, this was done according to the Pedigree-Matrix. For each row in Table A. 7 one column had to be chosen. Based on the resulting vector the total uncertainty for a flow was determined according to Equation A. 70. If a base uncertainty was available, it needed to be implemented as geometric standard deviation ( $\sigma_{base}$ ).

$$\sigma_{g,tot} = \exp \sqrt{\ln(\sigma_{g,1}^2) + \ln(\sigma_{g,2}^2) + \ln(\sigma_{g,3}^2) + \ln(\sigma_{g,4}^2) + \ln(\sigma_{g,5}^2) + \ln(\sigma_{base}^2)}$$
 Equation A. 70

Table A. 7: Pedigree-Matrix according to the ecoinvent data quality system

	_	•	•	4	_
	1	2	3	4	5
Reliability	Verified data based on meas- urements	Verified data partly based on assumptions or non-verified data based on meas- urements	Non-verified data partly based on qualified esti- mates	Qualified esti- mate (e.g. by in- dustrial expert)	Non-qualified es- timate
$\sigma_{g,1}$ =	1	1.05	1.1	1.2	1.5
Complete- ness	Representative data from all sites relevant for the market considered over an adequate period to even out normal fluctuations	Representative data from >50% of the sites rele- vant for the mar- ket considered, over an ade- quate period or even out normal fluctuation	Representative data from only some sites (<<50%) rele- vant for the mar- ket considered or >50% of sites but from shorter periods	Representative data from only one site relevant for the market considered or some sites but from shorter pe- riods	Representative- ness unknown or data from a small number of sites and from shorter periods
$\sigma_{g,2}$ =	1	1.02	1.05	1.1	1.2
Temporal cor- relation	Less than 3 years of differ- ence to the time period of the da- taset	Less than 6 years of differ- ence to the time period of the da- taset	Less than 10 years of differ- ence to the time period of the da- taset	Less than 15 years of differ- ence to the time period of the da- taset	Age of data un- known or more than 15 years of difference to the time period of the dataset
$\sigma_{g,3}$ =	1	1.03	1.1	1.2	1.5
Geographical correlation	Data from area under study	Averaged data from larger area in which the area under study is in- cluded	Data from area with similar pro- duction condi- tions	Data from area with slightly simi- lar production conditions	Data from un- known or dis- tinctly different area
$\sigma_{g,4}$ =	1	1.01	1.02	1.05	1.1
Further tech- nological cor- relation	Data from enter- prises, pro- cesses and ma- terials under study	Data from pro- cesses and ma- terials under study (i.e. identi- cal technology) but from different enterprises)	Data from pro- cesses and ma- terials under study but from different technol- ogy	Data on related processes or materials	Data on related processes on la- boratory scale or from different technology
$\sigma_{g,5}$ =	1	1.05	1.2	1.5	2

# Appendix P. Inventoried data for the economic assessment

In Table A. 8 the economic lifetimes and background data for calculating the WACC are given. To account for economic fluctuations, the parameters that would affect the capital costs (the rates of interest and inflation) were varied by ±50%. As an Ex Ante approach was chosen,

innovative technologies were not subject to a higher risk potential and thus this was not reflected in the WACC. The background data on interest rates and shares of equity and debt capital were derived from KOST ET AL. (2018). "Larger [power] plants, constructed and operated by large investors and institutions have a higher WACC due to the expected return of the investor compared to small and medium size projects." (KOST ET AL. 2018). Thereof, centralized plants were impinged by higher shares of equity and interest rates. The rate of inflation was set to i=2% throughout. To account for economic fluctuations, the coefficients i, rec and rpc were varied by ±50% (triangular distribution).

Table A. 8: Economic lifetimes (It), share of equity and debt capital (EC, DC), respective interest rates ( $r_{EC/DC}$ ) and referenced throughput.

	lt	EC	DC	rec	r <sub>DC</sub>	throughput
biogas plant	20	20%	80%	8%	4%	
composting plant	20	20%	80%	8%	4%	
HTC-plant	15	20%	80%	8%	4%	dan an substrate and
Gasification plant (fixed bed/fluidized bed)	20	20%	80%	8%	4%	dep. on substrate and treatment path
Pyrolysis plant	20	20%	80%	8%	4%	
belt dryer	20	20%	80%	8%	4%	
lignite power plant	35	40%	60%	11%	5.5%	3,150,000 t FM
waste incineration plant	30	40%	60%	11%	5.5%	240,000 t FM
mono incineration plant	30	40%	60%	11%	5.5%	40,000 t DM
WWTP	20	20%	80%	8%	4%	600,000 PE

In Table A. 9 the implemented investment and O+M costs are summarized. For P-recovery from sewage sludge only treatment costs were available. They were implemented as O+M costs and in alignment to MONTAG ET AL. (2015) they were 5-15€/kg P<sub>recovered</sub>. When digestate from OFMSW and food waste were spread, revenues for digestate were calculated from the fertilizer value (0.76 €/kg N<sub>creditable</sub>, 0.64 €/kg P2O5, 0.62 €/kg K2O and 0.17€/kg C<sub>Humus</sub>). The shares of the nutrients were taken from the elemental composition.

If external heat was required in the systems, the net natural gas prices for industries in Germany, for the period between 2008 and 2018, were taken as reference for the specific gas price (3.9±0.5 ct/kWh) (BMWI 2019).

The initial waste collection costs of OFMSW and food waste were 84.6€/t ± 50% (KRANERT 2017) and 25€/t ± 50% for the transport to the first treatment plant (door-to-door collection). For 'I', source segregation of food waste and OFMSW was not required, and the collection costs were reduced according to the shares of the substrates in the total residual waste. This was considered as a compromise between not accounting for the waste collection and a full integration of the collection costs. For waste wood, source segregation was always assumed, as the referenced waste wood was considered as hazardous waste. The logistics associated with waste wood collection are significantly different from those of household waste, but due to lack of data the costs could only be roughly estimated from waste statutes of municipalities. A value of 10€ ± 50% was used.

The transportation distance to a waste incineration plant was 40 km and costs were assumed to be 25-35 €/t for an intermediate transport between a decentralized plant and a municipal waste incineration unit. For the transportation of sewage sludge to a centralized incineration unit (distance=50 km) the costs were 30-40 €/t. If material was transported from the point of waste collection or the first treatment plant to a lignite power plant, transportation costs of 50-

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80 €/t were assumed. Transportation of residues was already accounted for in the respective O+M costs of the treatment plants.

Table A. 9: Distributions for net Investment and Operation and maintenance costs for the considered components. Operation and maintenance costs do not contain

Component	referenced	l₀ [Mio €]ª	(O+M)t [Mio €]	data basis (upscaled to referenced throughput)
	throughput [t/a]			
				I <sub>0</sub> = 9,052,387 €; O+M= 812,008 € (BALUSSOU ET AL. 2012)
		triangular: min= 4.2 mode= 10.5 max=18.4	triangular: min= 0.41 mode= 0.77	I <sub>0</sub> = 4,843,159 €; O+M= 411,668 € (KARELLAS ET AL. 2010)
biogas plant, dry				I <sub>0</sub> = 18,357,597 €; O+M= 848,801 € (Staiß et al. 2007)
fermentation (incl.	40.000 t FM/a			I <sub>0</sub> = 4,219,417 €; O+M= 524,530€ (FACHAGENTUR FÜR NACHWACHSENDE ROHSTOFFE 2013)
CHP plant)			max=1.22	$I_0$ = 10,610,016 $\in$ ; O+M= 1,228,000 $\in$ (Kern et al. 2010)
		max-10.1	max=n22	I <sub>0</sub> = 15,840,000 (RAUSSEN AND SPRICK 2012)
				O+M=812,000 (DEUTSCHE VEREINIGUNG FÜR WASSERWIRTSCHAFT, ABWASSER UND ABFALL
				2015)
				I <sub>0</sub> = 4,682,635 €; O+M=420,037 € (BALUSSOU ET AL. 2012)
				I <sub>0</sub> = 2,000,000 €, O+M=170,000 € (BALUSSOU ET AL. 2012)
bis as a slead west				I <sub>0</sub> = 2,497,613 €; O+M= € (KARELLAS ET AL. 2010)
biogas plant, wet fermentation	10.000 t FM/a	normal: $\bar{x}$ = 4.3	normal: $\bar{x}$ = 0.28	I <sub>0</sub> = 3,808,760 €; O+M= 395,852 € (STAIß ET AL. 2007)
(incl. CHP plant)	10.000 t FIVI/a	x = 4.3 $\sigma = 1.6$	x = 0.26 $\sigma = 0.1$	I <sub>0</sub> = 1,528,500 € Thünen-Institut (2010)
(IIICI. CI IF plant)		0= 1.0	0= 0.1	I <sub>0</sub> = 2,182,628 €; O+M=210,008 (FACHAGENTUR FÜR NACHWACHSENDE ROHSTOFFE 2013)
				I <sub>0</sub> = 4,618,277 €; O+M=197,926 € (KERN ET AL. 2010)
		triangular:		
		min: 1.7	+30% of biogas	I <sub>0</sub> = 1,716,810 € (RAUSSEN AND SPRICK 2012)
composting plant	28.600 t FM/a	mode: 2.5	plant	I <sub>0</sub> = 1,774,037 € (BILITEWSKI AND HÄRDTLE 2013)
		max: 4.2	piant	I <sub>0</sub> = 4,270,924 € (Hogg 2002)
				$I_0$ = 136,019,606 $\in$ ; O+M=12,865,441 $\in$ (Hogg 2002)
waste incineration		normal:	uniform:	O+M=12,940,967 € (FEHRENBACH ET AL. 2007)
plant	240.000 t FM/a	$\bar{x}$ = 101.6	min= 12.9	I <sub>0</sub> =68,750,492 €; I <sub>0</sub> =102,085,184 (RICHERS 2010)
L		$\sigma$ = 23.8	max= 13.0	I <sub>0</sub> =99,433,621 € (BILITEWSKI 2009)

Mono incineration plant for sewage sludge	40,000 t DM/a	triangular min: 41.6 mode: 59.4 max: 77.2	triangular min: 6.4 mode: 7.5 max: 8.6	I <sub>0</sub> = 136,019,606 €; O+M=12,865,441(NEBOCAT 2013)
HTC-plant (incl. post-treatment)	15.000 t DM/a	normal: $\bar{x}$ = 6.8 $\sigma$ = 3.3 uniform: min=6.1	triangular: min= 0.5 mode= 1.1 max= 1.9 triangular: min=0.68	$I_0$ =6,825,059 € (Wirth et al. 2011), $I_0$ =4,805,835 €; O+M=1.883,440 € (Erlach et al. 2011) $I_0$ =1,839,967 €; O+M=532,695 € (Lucian and Fiori 2017) $I_0$ =9,619,045 € (Stemann et al. 2013a) $I_0$ =10,941,707 €; O+M=960,470 € (Suwelack et al.) $I_0$ =6,112,714-13,719,647, O+M=98,100-1,297,100 (Shackley et al. 2011) O+M=1,349,649 (Roberts et al. 2010)
Pyrolysis	10,000 t FM	min=6.1 max=13.7	mode=0.99 max=1.35	O+M=1,235,343 (ABC ADVANCED BIOMASS CONCEPTSGMBH 2018) O+M=683,234 (RINGER ET AL. 2006)
Gasification plant (fixed bed)	500 kW <sub>el</sub>	triangular: min=3.7 mode=5.6 max=7.7	triangular: min=0.27 mode=0.53 max=0.89	I <sub>0</sub> =1,484,446 (C.A.R.M.E.N. E.V. 2016) I <sub>0</sub> =910,034, O+M=5-9% of I <sub>0</sub> (Zeymer et al. 2013) I <sub>0</sub> =1,370,565 (Strzalka et al. 2017) I <sub>0</sub> =1,825,000 (Kralemann 2014)
Gasification plant (fluidized bed)	1,000 kWel	triangular min=8.4 mode=15.1 max=23.8	triangular: min=0.4 mode=1.3 max=2.8	$\label{eq:loss} $I_0$=$12,716,030 (Strzalka et al. 2017)$                                    $
lignite power plant	3,150,000t char (DM=90%)/a	uniform: min= 1,208 max=1,785	57.2	$I_0 = 1,785,000,000 \in ; O + M = 57.225,000 \in (Hauke \ et \ al. \ 2017)$ $I_0 = 1,207,500,000 \in (Wissel \ et \ al. \ 2008)$
WWTPb	600,000 PE	triangular: min=16.9 mode=24.2 max=31.4	2.14	(BOLLE AND PINNEKAMP 2011; THÜNEN-INSTITUT 2010; BALUSSOU ET AL. 2012; IINAS 2020; ASUE 2011; FRICKE, MUTZ 2018)
Pastuerization unit at WWTP	600,000 PE	0.35	0.04	(Pospiech et al. 2014)

Costs for agricul- tural utilization of sewage sludge	1 t DM	-	normal: $\bar{x}$ = 3.44E-4 $\sigma$ = 6.9E-5	(HILMER 9/11/2019)
belt dryer	10.000 t of evaporated water	2.7	0.11	(HEINDL 2016)

a) extended by the replacement of components and - if not included - the costs for contingency, offsite costs, startup costs etc. b) incl. thickening, digestion, CHP, emergency flare, dewatering and dewatering

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