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Efficient Carbon, Nitrogen and Phosphorus cycling in the European Agri-food System and related up- and down-stream processes to mitigate emissions



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D3.1. Classification of food waste and wastewater streams in food industry and their recycling potential for carbon, nitrogen and phosphorus

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Author(s)	Anne Kleyböcker, Fabian Kraus, Wim Moermann, Natalia Pudova, Marek Holba, Andreas Dünnebeil
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Reviewed by	Christian Remy (KWB), Joachim Clemens (SOE) , Victor Riau (IRTA),
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Contact person EC	Johanna Schepers

Contributing partners	
1.	KWB
2.	NRS
3.	ASIO
4.	PON

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1 Introduction: background and objective

Circular Agronomics, aims to foster the transition from a linear economy to a circular economy. Therefore, this deliverable focuses on circular solutions for waste and wastewaters originating from the food industry. In 2019, the “Best Available Techniques (BAT) Reference Document for the Food, Drink and Milk Industries” (BREF-document) was published by the European Commission. Based on that, the deliverable summarizes the state of the art of the technologies already in use and concludes their suitability for circular economy solutions.

In Circular Agronomics, new technologies for the recovery of carbon, nitrogen, phosphorus and potassium are developed and investigated. So far, those technologies are not included in the BREF-document yet. Therefore, the concepts of the technologies are introduced in the deliverable. For a potential integration of those technologies in the BREF-document, the technologies are described in detail in the annex according to the required structure in the BREF-document. However, since the technologies are still under development, those descriptions are considered as a first draft. The authors suggest to update those descriptions at a later stage of the project prior to their potential integration in the BREF-document.

Referring to the goal to recover carbon and nutrients, the deliverable presents a detailed characterization of the waste and wastewaters originating from the food industries. Based on that, the five most promising waste and wastewater streams regarding carbon recovery, nitrogen recovery, phosphorus recovery and potassium recovery were selected. For those streams and the corresponding recovery technologies four new concepts are suggested in the deliverable. In order to show the technology providers an overview of potential clients for their technologies and for those concepts, for each selected industry, the European country with the highest production rate was chosen. For this country, the regional distribution of the certain industry was determined.

2 State of the art: review of best available techniques (BAT) for the removal of C, N, and P from food and drink processing wastewater

In the BREF-document (2019), best available techniques for wastewater treatment in the food, drink and milk industries are summarized and listed. A short review of those technologies for the three categories carbon, nitrogen and phosphorus removal, treatment and recovery is presented in the following.

2.1 BAT for C removal, treatment and recovery

For carbon removal, this deliverable distinguishes between mechanical carbon removal, aerobic carbon degradation and anaerobic carbon degradation.

2.1.1 Mechanical carbon removal from wastewater and/or thickening

In the BREF-document (2019), different technologies are listed for the mechanical separation of solids from wastewater. Especially for small particles resulting from fruit or vegetable processing up to coarse solids, different **types of screens** are recommended as BAT depending on the opening size between the screen bars or the gaps of the perforated plates (Fig. 1).

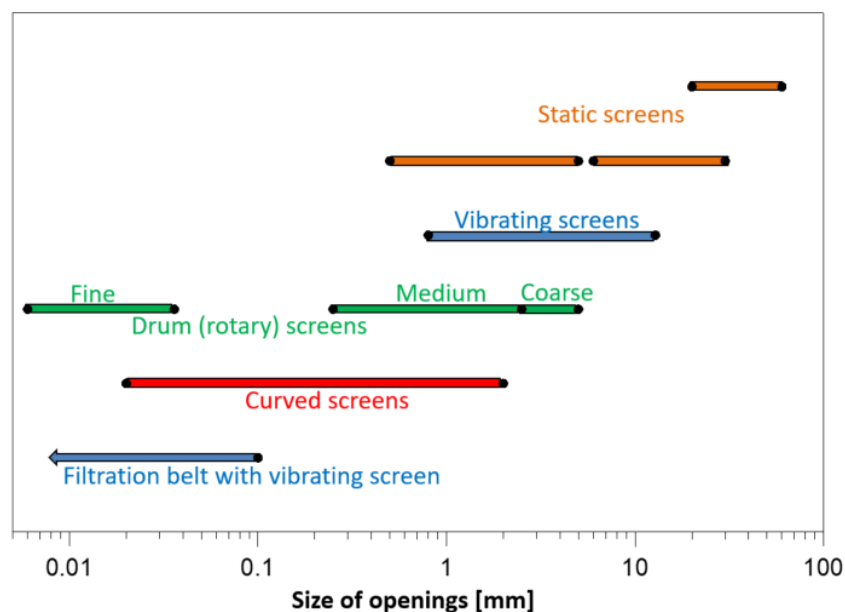


Fig. 1 Summary of different screen types depending on the openings or gaps widths between the bars or in perforated plates

Those are static screens with openings of up to 70 mm, vibrating screens with a maximal opening of 15 mm, curved screens with 2mm and smaller openings, rotary drum screens ranging from 6 μm to 5 mm and the combination of a filtration belt with a vibrating screen with openings that are smaller than 0.1 mm.

For the mechanical separation of smaller particles with a diameter of 10 μm and less, **membrane filtration processes** must be applied. In Fig. 2, the BAT listed in the BREF-document are summarized and presented depending on the pore size of the filter or membranes. Hereby, especially for filtration techniques in the range of ultrafiltration and also with smaller pore sizes, a pretreatment and/or cross-flow instead of dead end filtration is recommended in order to prevent fouling.

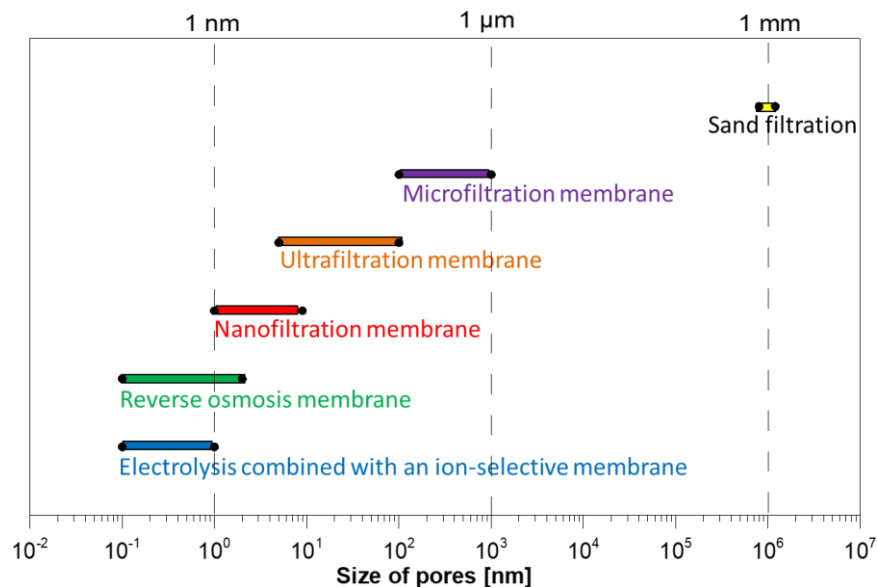


Fig. 2 Summary of different BAT for filtration depending on the pore sizes of the membrane or the sand filter (the ranges were defined according to Gujer 2007 and Razmjou et al. 2019)

For the pretreatment such as the **removal of fat, oil and grease (FOG)** as well as for light hydrocarbons, BATs are according to the BREF-document:

- Fat trap/grease interceptor
- Parallel-plate separator with a plate angle of 45° (prone to clogging due to vegetable oils)
- Combination of augers and flotation tanks
- Dissolved air flotation by injecting fine air bubbles in a floating reactor (80% FOG removal)
- Mechanical removal of the floating material via skimming or suction withdrawal (80% FOG removal)
- Flotation combined with coagulation and flocculation (<95% FOG removal)

In contrast to the flotation technologies, suspended solids, flocs and precipitates with a higher density than water can also be easily separated via **sedimentation**. Therefore, the BREF-document lists:

- Rectangular tanks
- Circular tanks
- Laminar separators (prone to blockages with fat)

Suspended solid particles can be so small, that they cannot be separated via sedimentation. Those particles as well as dissolved substances can also be removed via **coagulation and flocculation**. Therefore, additives such as $\text{Al}_2(\text{SO}_4)_3$, FeCl_3 and CaO are used for coagulation and polyelectrolytes for flocculation. Subsequently, the resulting compounds are removed via sedimentation or flotation depending on their density.

Conclusion for CIRCULAR ECONOMY solutions: most of the technologies are SUITABLE

Especially, in terms of a subsequent reuse option of the separated material, technologies which do not involve any addition of chemicals are suitable in particular. Thus, coagulation and flocculation should be only applied, if there is a reuse option

for which the additional substances are acceptable. Otherwise, it is recommended to remove the particles or dissolved substances only via a mechanical separation without the addition of further substances.

2.1.2 Carbon removal via aerobic degradation

Soluble organic carbon compounds are often removed from wastewater via aerobic degradation. In this process, aerobic heterotrophic microorganisms degrade dissolved organic carbon compounds to CO₂ and H₂O using oxygen (Madigan et al. 2012). Furthermore, the microorganisms use the carbon to grow and thus, form biomass. Mainly two different systems can be distinguished. The activated sludge system in which microorganisms are suspended usually in the form of flocs in the wastewater and fixed biofilm aerobic systems with immobilized microorganisms. In the BREF-document (2019) different technologies based on the **activated sludge system** are listed:

- Conventional activated sludge system with an aeration tank and a settling tank
- Aerobic lagoons which are periodically mixed with pumps or mechanical aeration
- Pure oxygen systems consisting of a conventional system operated with pure oxygen injection instead of air, in the case, the plant exceeds temporarily its foreseen capacity for air aeration
- Sequencing batch reactors in which aeration and settling take place in succession in the same reactor
- Aerobic membrane bioreactor with a submerged membrane or an external membrane

Those activated sludge systems are summarized depending on their organic loading rate and their food to microorganism ratio in Fig. 3 and Tab. 1.

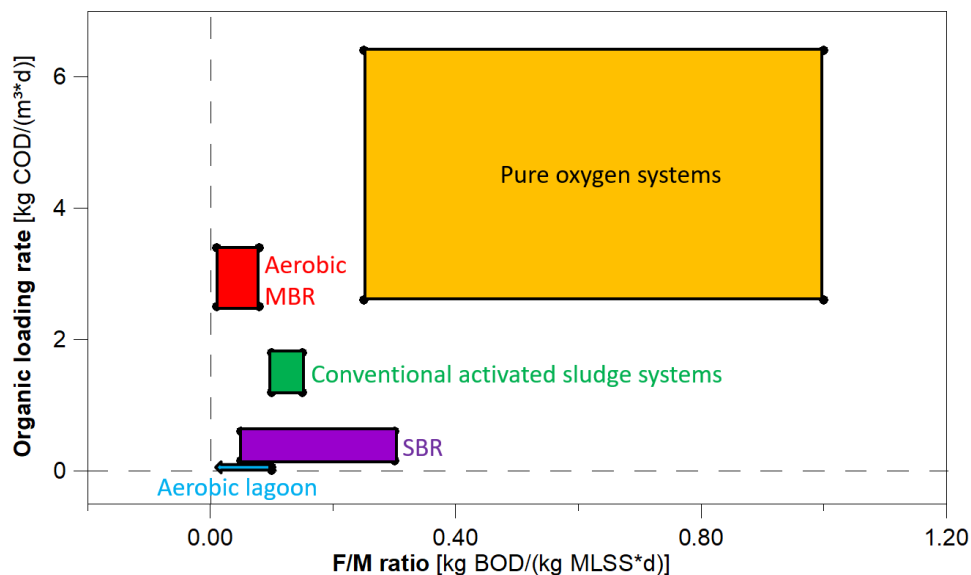


Fig. 3 Summary of activated sludge systems depending on their F/M (food/microorganism) ratio and their organic loading rate according to Tab. 1

Tab. 1 Food to microorganism (F/M) ratios and organic loading rates (OLRs) of different activated sludge systems

Process/technology	F/M ratio [kg BOD/(kg MLSS*d)]	OLR [kg COD/(m ³ * d)]	Literature reference
Conventional activated sludge system	0.1 – 0.15	1.2 – 1.8	BREF 2019
Aerobic lagoons	Low	0.01 – 0.05	Lorch 1996, Metcalf et al. 1991
Pure oxygen systems	0.25 – 1	2.6 – 6.4	BREF 2019, Metcalf et al. 2013
Sequencing batch reactors (SBR)	0.04 – 0.3	0.16 – 0.6	Metcalf et al. 2013, Fernandes et al. 2013; Lefebvre et al. 2005
Aerobic membrane bioreactor (MBR)	0.01 – 0.08	2.5 - 3.4	Delgado et al. 2011, Bracklow 2012; Wang et al. 2005, Vocks 2008

Unfortunately, in those systems, the aeration via air injection usually consumes around 50% or even more of the energy demand of a full-scale wastewater treatment plant depending on its size and the employed technological solutions (Drewnowski et al. 2019). The CO₂ resulting from the aerobic degradation process is usually stripped via the aeration from the treated wastewater and emitted into the air. The produced biomass must be removed as excess sludge from the liquor. Activated sludge systems produce high amounts of excess sludge which need to be disposed. Therefore, the excess sludge is usually thickened and valorized as substrate for biogas production. Thus, the CO₂ is lost, but at least the biomass of the excess sludge can be reused for energy production. However its biodegradation is usually only partially achieved (typically <50%) in a biogas production process.

For the **fixed biofilm systems without air injection**, the BREF-document defines the following technologies as BAT:

- Trickling filters: non-submerged aerobic fixed biofilm reactors, that use rock or plastic packing over which wastewater is distributed for treatment
- Bio-towers: trickling filters for higher organic loading rates in above-ground tanks containing plastic media for a higher specific surface area
- Rotating biological contactors (combination of floc and biofilm based processes)

Compared to the activated sludge systems, the fixed biofilm systems produce less excess sludge. They do not need energy for aeration, because the oxygen of the surrounding air is used by the microorganisms for the aerobic organic carbon degradation. Trickling filters are often used as posttreatment or polishing step, while bio-towers can treat higher organic loading rates and hence, are frequently used as pretreatment (Tab. 2). However, besides the rotating biological contractor, all listed biofilm systems are prone to blockages. Thus, coarse solids should be removed from the wastewater prior to the treatment.

For the **fixed biofilm systems with additional aeration** by air injection via nozzles the BREF-document defines high and ultra-high rate filters as BAT. Due to the better availability of oxygen in the system, because of its artificial aeration, those filters can also treat high organic loading rates (Tab. 2) and, hence are also often used as a pretreatment technology.

Tab. 2 Overview on the BAT for aerobic carbon degradation in fixed biofilm systems

Technology	OLR [kg COD/(m ³ * d)]	Literature
Trickling filters	0.4 – 0.8	Gujer 2015
Bio-towers	1 – 5	BREF 2019
Rotating biological contractors	1.2 – 2*	Cortez 2008; *calculated from OLR (referred to the surface) multiplied with the density (=surface to reactor volume).
High and ultra-high rate filters	1.2 – 4.8	Metcalf et al. 2013

Conclusion for CIRCULAR ECONOMY solutions: only SUITABLE as a polishing step

Those technologies are not suitable as a key technology for fostering circular economy solutions, since they do not recover nutrients nor produce energy or carbon-rich material. Excess sludge in form of biomass is low in quantity and relatively hard to degrade and convert into biogas. In contrast, the aerated systems even consume a lot of energy. The degradation end product from organic carbon compounds is CO₂, which is emitted into the air. However, as already mentioned, for a polishing step in a posttreatment to reach a good water quality, they can be considered.

2.1.3 Carbon removal via anaerobic degradation

For the organic carbon removal via anaerobic degradation, different technologies are listed as BAT in the BREF-document (2019). The technologies differ between wash out systems and systems with biomass accumulation. The **anaerobic contact process** is a typical wash out system and takes place in continuously stirred tank reactors. Herein, the biomass is distributed in the form of flocs within the reactors. The solids retention time and the hydraulic retention time are equal. However, in systems with biomass accumulation, the biomass is kept in the reactor via pellets, as biofilm on package

material in fixed or fluidized bed systems or as flocs in an anaerobic membrane system. Here, the solids retention time is decoupled from the hydraulic retention time. According to the BREF-document, following systems based on pellets are defined as BAT:

- **upflow anaerobic sludge blanket reactors (UASB),**
- **internal circulation reactors** consisting of two UASB reactors
- **expanded granular sludge bed reactors.**

For fixed or fluidized bed systems, two types are indicated as BAT:

- **anaerobic filters**, which contain packing material with immobilized microorganisms and which can be operated in both directions, upflow or downflow and
- **fluidized and expanded bed reactors**, in which the packing material with immobilized microorganisms is hovering due to a higher upflow velocity than in an anaerobic filter

Furthermore, an **anaerobic membrane bioreactor** is mentioned as BAT as well. It is especially suitable for the degradation of more refractory organic materials, because the biomass retention time can be adjusted as long as necessary for a successful degradation process. However, one disadvantage is, that the membranes have to be cleaned periodically with chemicals.

In Tab. 3, the technologies are listed with their typical organic loading rates. While the wash out systems can only handle relatively low organic loading rates (OLR) with 5 kg COD/(m³ * d) and below, the systems with biomass accumulation can be operated with up to 35 kg COD/(m³ * d).

Tab. 3 Overview on the BAT for anaerobic carbon degradation

Process/technology	OLR [kg COD/(m ³ * d)]	Reference
Anaerobic lagoons	<1 – 2	Deublein et al. 2008
Anaerobic contact processes	≤ 5	BREF 2019
Anaerobic filters	< 12	Deublein et al. 2008
Fluidized and expanded bed reactors	15 – 35	BREF 2019
UASB	5 – 15	BREF 2019, Rosenwinkel et al. 2015
Internal circulation reactors	15 – 35	BREF 2019
Expanded granular sludge bed reactors	15 – 25	BREF 2019
Anaerobic membrane bioreactor	2 – 15	Rosenwinkel et al. 2015

The benefit of anaerobic organic carbon degradation is, that the carbon is converted to methane and CO₂. The methane can be recovered and further reused as an energy source. This is true for all listed systems, besides the anaerobic lagoon. The anaerobic lagoon is indicated as BAT. However, it does not allow for the collection of the produced biogas and thus, the biogas is emitted into the air contributing to the undesired greenhouse effect. The collection and recovery of dissolved methane from anaerobic reactor effluents is a challenge and requires specific technologies for degassing to maximize methane recovery and prevent unwanted losses of this greenhouse gas to the atmosphere.

Furthermore, depending on the substrate, biomass generation in anaerobic systems is between 3- and 10-fold lower than in aerobic systems (Rosenwinkel et al. 2015). This is also an advantage regarding the costs for a potential sludge or biosolids disposal.

Anaerobic digestion systems are also suited for co-digesting wastewaters with high organic loads together with organic wastes (see also chapter 4). Depending on the substrate composition, phosphate and ammonium compounds can result from the fermentation of more complex organic compounds. Thus, the effluent resulting from the anaerobic treatment needs further treatment, before it can be released into the environment. Moreover, the availability of ammonium and phosphate allows for their recovery (see also chapter 3).

Conclusion for CIRCULAR ECONOMY solutions: SUITABLE

Anaerobic digestion is very suitable for circular economy solutions, because it recovers energy from carbon substrates in form of biogas and produces a largely stabilized organic material. Especially for the food industry, the joint treatment of its wastewaters with its organic wastes via co-digestion is very promising leading to high methane yields (see chapter 4). In

this case, due to the microbial degradation processes in the anaerobic reactor, in addition to biogas, also organic bound nitrogen and phosphorus are converted into ammonium and phosphate. Thus, the effluent contains high concentrations of ammonium and phosphate, that is very suitable for nutrient recovery technologies as further presented in detail in chapter 3 and 5. Once, the digestate is dewatered it is also very suitable as a soil conditioner.

2.2 BAT for N removal, treatment and recovery

For nitrogen removal from wastewater, different technologies based on biological processes such as nitrification, denitrification and ANAMMOX or physical processes such as ammonia stripping are listed in the BREF-Documents (2019).

2.2.1 Nitrogen removal via nitrification and denitrification

Nitrogen removal via nitrification and denitrification are biological processes. During nitrification, aerobic autotrophic microorganisms oxidize ammonium to nitrate. In a subsequent anoxic process, heterotrophic microorganisms reduce the nitrate to molecular nitrogen. Hereby, the heterotrophic bacteria consume organic compounds and produce CO₂ in addition to the molecular nitrogen. The molecular nitrogen is emitted into the air as nitrogen gas. Thus, there are no reuse options for the removed nitrogen from the wastewater. The BREF-document does not list any specific technologies for those processes in the corresponding chapter. However, in the following the most common technologies, which can be designed to contain nitrification and denitrification stages, are listed:

- Combination of aeration tank (AE) for carbon degradation and nitrification as well as a continuously stirred tank reactor for denitrification (AX) (upstream or downstream of the aeration tank for pre- and post-denitrification, respectively)
- Sequencing batch reactor with the configuration of an aeration time period and an anoxic time period
- Membrane bioreactor including the combination of AE and AX

Conclusion for CIRCULAR ECONOMY solutions: NOT SUITABLE

Those technologies are not suitable for fostering circular economy solutions in terms of nutrients or energy recovery, since they do not recover them. The degradation products are nitrogen gas and CO₂ which are both emitted into the air.

2.2.2 Nitrogen removal via ANAMMOX

The nitrogen removal via ANAMMOX is a biological process as well as the nitrification and the denitrification processes. However, the difference to those processes is, that during nitritation, the ammonium is partially oxidized to nitrite only with around 50% of the ammonium converted to nitrite (Koch et al. 1998). In a subsequent anaerobic ammonium oxidation (ANAMMOX), the remaining 50% of the ammonium is oxidized under anoxic conditions. Hereby, the nitrite is used as electron donor and molecular nitrogen and water are formed. Thus, compared to the nitrification and denitrification, the ANAMMOX processes need 60% less oxygen (Mudrack et al. 2003) and no organic carbon at all, because the relevant microorganisms are autotrophic. Furthermore, the excess sludge production comprises only 10% of that resulting from the conventional nitrification/ denitrification (Mudrack et al. 2003). The BREF-document (2019) does not list any specific technologies for those processes in the corresponding chapter. However, in the following, typical technologies for nitritation and anaerobic ammonium oxidation are listed according to Rosenwinkel (2015) and Trigo et al. (2006):

- Combination of aeration tank (AE) for carbon degradation and partial nitritation as well as a continuously stirred tank reactor for the subsequent ANAMMOX (AX)
- Sequencing batch reactor with aeration time periods and anoxic time periods (either one-stage or two-stage configuration)
- Membrane sequencing batch reactor with aeration time periods and anoxic time periods
- Airlift reactor with granulated sludge
- Moving bed reactor containing different zones with aerobic and anoxic conditions

Conclusion for CIRCULAR ECONOMY solutions: NOT SUITABLE

Those technologies are not suitable for fostering circular economy solutions, since they remove the nitrogen, but they do not recover any nitrogen. The end product is nitrogen gas which is emitted into the air. However, compared to the nitrification and denitrification processes, the energy demand of the ANAMMOX process is much lower and it does not consume organic carbon for the nitrogen removal.

2.2.3 Nitrogen removal via ammonia stripping

The nitrogen removal via ammonia stripping is a physical process. For this process, the equilibrium between ammonium and ammonia has to be shifted towards the gaseous ammonia side. Therefore, the pH and the temperature are crucial. The higher the pH and temperature are, the more that equilibrium shifts to the ammonia side. Therefore, the wastewater is usually alkalized with sodium hydroxide to a pH of 9 and higher and enters a desorption column. This column is filled with packing material in order to increase the water air interface. The wastewater flows downwards the column, while the air is injected at the bottom of the column and is circulated upwards through the column. In doing so, the ammonia is released from the liquid phase into the gaseous phase. Subsequently, the gaseous phase containing the released ammonia passes through a gas scrubber containing sulfuric acid. Here, the ammonia reacts with the sulfuric acid to ammonium sulfate. Currently, only ammonia stripping via air injection with a subsequent ammonia gas scrubber containing sulfuric acid is defined as BAT in the BREF-document (2019). However, there exist more technologies for ammonia stripping which the authors recommend to include in the BREF-document. Therefore, in chapter 3.2 those technologies will be described in detail.

Conclusion for CIRCULAR ECONOMY solutions: SUITABLE

Ammonia stripping technologies are suitable for fostering circular economy solutions, since they do not only remove ammonia, but also recover it for a further processing to for example ammonium sulfate being a typical nitrogen fertilizer. See also chapter 3.2 for more information.

2.3 BAT for P removal, treatment and recovery

In the food, drink and milk industry, chemical processes in order to precipitate phosphorus compounds and remove them from the wastewater are currently considered as BAT. Furthermore, also enhanced biological phosphorus removal which is based on biological processes is listed as BAT.

2.3.1 Chemical phosphorus removal via precipitation

For chemical phosphorus removal, usually certain chemicals such as iron salts or solutions are mixed into the wastewater. The chemicals precipitate with the dissolved phosphate contained in the wastewater e.g. as iron phosphate. Subsequently, the phosphate precipitates are mechanically separated from the wastewater and thus, the phosphorus is removed. Typical chemicals in order to induce the precipitation of phosphorus compounds are:

- FeCl_2 , FeCl_3 , FeClSO_4 or FeSO_4 in order to precipitate iron phosphate compounds such as vivianite
- $\text{Al}_2(\text{SO}_4)_3$ or $\text{NaAl}(\text{OH})_4$ in order to precipitate aluminum phosphate compounds such as berlinite
- MgCl_2 , MgO or $\text{Mg}(\text{OH})_2$ in order to precipitate struvite for example (see 2.3.3 *struvite precipitation*)
- CaO or $\text{Ca}(\text{OH})_2$ to precipitate calcium phosphate compounds such as hydroxyapatite or brushite

In the BREF-document (2019) a precipitation system is mentioned as BAT working with lime and consisting of four tanks: a softening tank, a coagulation tank, a flocculation tank and a settling tank with lamellae and a scraper. In the coagulation tank, the lime is mixed into the wastewater for the phosphorus precipitation. In the flocculation tank, microsand is added in order to accelerate the settling process in the fourth tank. The microsand is later on separated via a hydrocyclone and thus, can be reused again in the process.

Conclusion for CIRCULAR ECONOMY solutions: depending on the chemical type of P precipitate – SUITABLE (see chapter 3.3.)

As long as the precipitation product is suitable for recycling such as struvite, hydroxyapatite and brushite, this method for P removal is suitable to foster circular economy. However, the precipitation products resulting from iron or aluminum additives are not suitable for direct recycling, because iron phosphates as well as aluminum phosphates are not available for plants under normal pH conditions in soils (Desmidt et al. 2015). Precipitates from iron or aluminium addition need to be dissolved again in acidic solution, thus requiring a high amount of chemicals. In chapter 3 and 5, concepts are described in detail regarding phosphorus recovery in the form of struvite, K-struvite and brushite.

2.3.2 Enhanced biological phosphorus removal (EBPR)

The EBPR is based on the activity of so-called polyphosphate accumulating organisms (PAOs). Therefore, PAOs need to be enriched and activated by alternating process conditions (anaerobic, aerobic) to successfully remove phosphate from the wastewater. According to Seviour et al. (2003), under anaerobic conditions and with the availability of volatile fatty acids (VFAs), the PAOs release phosphate from their polyphosphate storage, take up VFAs and store them as poly-β-hydroxyalkanoates (PHAs). While under aerobic conditions, and if substrate is limited, they can use the stored PHAs and take up phosphate for growing. In addition, they even store excess phosphate as polyphosphates. Subsequently, the PAOs containing the stored polyphosphate are removed from the treated wastewater at the end of the aerobic phase with the excess sludge discharge. An advantage of this process is, that the addition of chemicals can be avoided. Furthermore, there is still the opportunity to easily recover the stored polyphosphate from the PAOs in a slightly acidic and anaerobic milieu. In the BREF-document the following technologies are listed as BAT for EBPR:

- **A/O process** for mainstream phosphorus removal including carbon oxidation: combination of a CSTR with anaerobic conditions (AN) containing short fatty acids and an aerobic tank (AE) in sequence
- **Phostrip process**: here, in addition to the EBPR with the subsequent excess sludge discharge, a side stream of the P rich excess sludge is thickened under anaerobic conditions in order to release the stored polyphosphate. In the liquor the released phosphate is precipitated and can be recovered as a P containing mineral.

However, much more variants of the processes as mainstream treatment exist

- incl. an additional anoxic stage (AX): Johannesburg process, ISAH process, Phoredox process, Biedenpho process etc. (Mudrack et al. 2003)
- membrane bioreactor with AN, (AX) and AE in sequence
- sequencing batch reactor with anaerobic, aerobic and anoxic time periods in sequence

Conclusion for CIRCULAR ECONOMY solutions: SUITABLE as a P removal process prior to the P recovery process

Enhanced biological phosphorus removal is suitable for fostering circular economy solutions, because the stored polyphosphate can be easily recovered from the PAOs via an anaerobic treatment containing VFAs such as anaerobic digestion. After the phosphate release and dewatering, the phosphate is available for a phosphorus recovery at a later stage in the treatment train.

2.3.3 Phosphorus removal and recovery as struvite

The BREF-document defines phosphorus removal via struvite precipitation as a BAT especially for wastewaters or process waters with PO₄-P concentrations of 50 mg/L and above (Cornel and Schaum, 2009). Even though struvite is the name of a mineral family, in the wastewater sector it is usually used as a name for magnesium ammonium phosphate (MgNH₄PO₄·6H₂O). It is a slow release fertilizer (Kratz et al. 2019) and all three nutrients are plant available as from mineral fertilizers (Watson et al. 2019). In order to precipitate struvite, the phosphorus has to be available as dissolved phosphate in the wastewater together with ammonia and magnesium. Therefore, a pretreatment such as anaerobic digestion or even a combination of anaerobic digestion with an additional hydrolysis such as a thermal pressure hydrolysis or a thermal alkaline hydrolysis can lead to an increase in the phosphate concentration to 60% or even more of the total phosphorus content (Wilken et al. 2019). In order to successfully precipitate struvite, a pH of 8.5 and higher is required. Hence, as a first step towards a higher pH, the CO₂ is stripped from the wastewater via air injection. In a second step, NaOH is added, if the CO₂ stripping has not reached the required pH value. As already mentioned in chapter 2.3.1 *Chemical phosphorus removal*, to induce struvite precipitation, together with a certain ammonium concentration, the magnesium as MgCl₂, MgO or Mg(OH)₂ is usually added. This takes place in a reaction tank, typically in a continuously stirred tank reactor. Crystal growth is promoted by mixing, sufficient retention time and recirculation of formed crystals. As a last step, the struvite in form of larger crystals is separated in a settling tank. Usually, the struvite is dewatered, dried and processed, before it can be applied as a slow-release fertilizer.

Variants of the process: liquor - sludge

If the CO₂ stripping and struvite precipitation take place **in the liquor** (e.g. after dewatering), the subsequent separation of the struvite is very efficient. However, the higher phosphate concentrations are, the lower the dewatering efficiency of

the upstream dewatering unit is (Kuhn et al. 2013). Thus, the dewatering step might require more energy and sometimes even additives such as polymers in order to reach the required liquor quality, typically with a maximum content of total suspended solids (TSS) of 600 mg/L (Ohl 2020). In the liquor, the crystals grow usually homogeneous.

If the CO₂ stripping and struvite precipitation take place **in the sludge**, the separation of the struvite crystals is less efficient and the crystals are usually inhomogeneous due to organic and/or inorganic impurities. However, the controlled struvite precipitation can be a useful measure to prevent pumps or pipes in the sludge line from scaling or even clogging (Desmidt et al. 2015).

Requirements and favorable conditions for struvite precipitation:

- pH: 7.5 – 9 (Lahav et al. 2013, Shaddel et al. 2019)
- PO₄-P concentration > 50 mg/L (Cornel and Schaum, 2009)
- Mg:N:P molar ratio: 1:2:1 – 1:12:1 (Shaddel et al. 2019)
- low activity based supersaturation with $S_a \leq 3.3$: stronger aggregation of struvite crystals (Shaddel et al. 2019)

Conclusion for CIRCULAR ECONOMY solutions: SUITABLE

Struvite recovery is very suitable for fostering circular economy solutions, since struvite is a slow release fertilizer and all nutrients such as P, NH₄ and Mg are as good plant available as the nutrients of a mineral fertilizer.

3 Circular economy (CE) technologies developed and investigated in Circular Agronomics

In Circular Agronomics, new technologies for the recovery of carbon, nitrogen, phosphorus and potassium are developed and investigated. So far, those technologies are not included in the BREF-document yet. Therefore, the concepts of the technologies are introduced in this chapter. For a potential integration of those technologies in the BREF-document, the technologies are described in detail in the annex according to the required structure in the BREF-document. However, since the technologies are still under development, those descriptions are considered as a first draft. The authors suggest to update those descriptions at a later stage of the project prior to their potential integration in the BREF-document.

In Fig. 4 an example is given for a circular economy concept comprising carbon, nitrogen and phosphorus recovery.

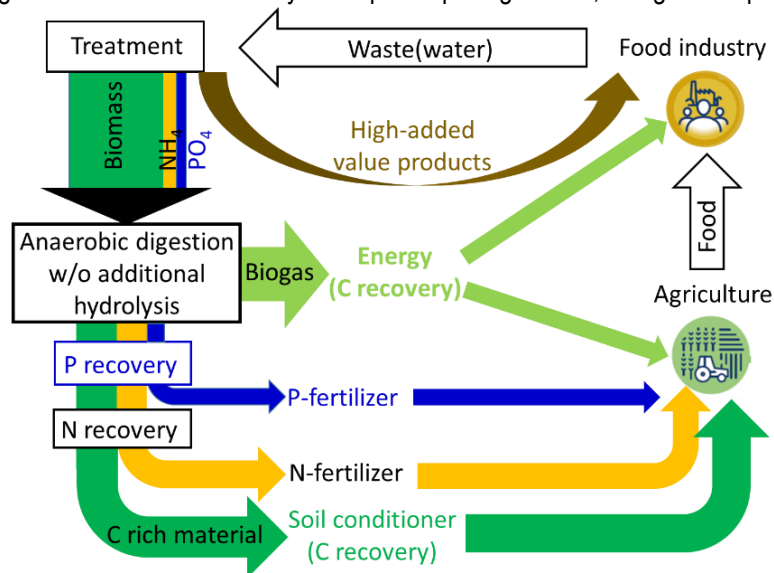


Fig. 4 Example for the carbon and nutrient cycle: from food to wastewater via recovery and agriculture back to food

Recovered products containing mainly carbon are for example high-added value products, substrate for biogas production, biogas, digestate and soil conditioner. Typical products resulting from nitrogen recovery are ammonia gas, ammonia water, ammonia nitrate, ammonium sulfate and ammonium carbonate. The last three compounds can be directly used as fertilizers. However, ammonia gas and ammonia water have to be further processed, if they shall be applied in agriculture. Typical products containing phosphorus are P-minerals such as (NH₄-)struvite, K-struvite, hydroxyapatite, brushite and tricalcium phosphate. Fig. 4 shows, that the combination of energy recovery and material recovery is very promising, because the biogas production process does not only recover energy, but it also contributes to the increase in the release of ammonium and phosphate from organic compounds being degraded via anaerobic digestion. The availability of nitrogen as ammonium and of phosphorus as phosphate is crucial for their recovery.

In the following chapters, the circular economy technologies being investigated in Circular Agronomics are presented in detail.

3.1 CE technologies for the recovery of C

Typical components in wastewaters from the food, drink and milk industry are carbon rich materials such as organic particles and solids, dissolved organic matter, FOG and excess sludge. Using different CE technologies, those components can be valorized as substrate for biogas production, from a few components high-added value products can be recovered and/or after a certain treatment the carbon rich material can be reused as soil conditioner.

For **high-added value products**, Ahmed et al. (2019) and Chen et al. (2019) give a comprehensive overview in their reviews on the products and the treatments in order to recover those products. Ahmed et al. (2019) for example considers 9 high-added value products from olive mill wastewater and Chen et al. (2019) distinguishes between 6 categories of high-added value products recovered from wastewater of the fruit and vegetable processing industry. Since this is a comprehensive topic and not investigated in Circular Agronomics, the authors of this deliverable will not further focus on this topic.

Depending on its source, organic carbon based material contains variable amounts of easily aerobically and anaerobically degradable compounds such as saturated, unsaturated and volatile fatty acids. Furthermore, complex compounds such as humic substances, cellulose and lignin are hardly degradable. While the degradable compounds are usually quickly degraded to CO₂ on farmland, hardly degradable compounds have the potential to increase C stocks. Thus, the reuse of those easily degradable substances as **biogas substrate** for anaerobic digestion prior to the application of the digestate on the fields has the benefit of recovering energy. In addition, the resulting digestate contains usually slowly degradable organic compounds and hence, it is very well suited as **soil conditioner** to increase the carbon content of the soil.

In this chapter, only the treatment trains which are investigated in Circular Agronomics are presented in detail. They comprise the production of substrate for biogas production and/or the treatment to gain a soil conditioner from acid whey.

3.1.1 Centrifugation and nanofiltration for acid whey thickening in order to produce a substrate for biogas production

In dairies and milk processing industries, whey results as the remaining liquid after milk has been curdled and strained. Whey is distinguished in acid whey and sweet whey. Usually sweet whey is a by-product from the hard cheese production and has several commercial uses. In contrast, acid whey is often considered as waste and is often discharged into the sewerage system as for example in Slovenia, where 150,000 t/a are disposed to the sewerage system (Zupančič Justin 2020). In order to valorize the acid whey, it is thickened for its reuse as biogas substrate and/or as soil conditioner. The valorization as biogas substrate is already practiced, however, the reuse as a soil conditioner is a new concept that will be investigated in detail in the project Circular Agronomics in work package 1.

Description of the treatment train or technology

The treatment train consists of two parts, a centrifugation unit and a nanofiltration unit. The nanofiltration unit is used for thickening of acid whey in order to increase the concentration of its macro components. As a filter, membranes from Dupont of the type DOW Filmtec NF 3840/30-F are used. They have a polypropylene outer shell and their pore size is approximately ~ 75 kDa. In order to protect the polymer membrane from fats contained in the feed whey, pre-centrifugation is used.

The centrifugation step reduces the fat content by 30%. Thickening of the acid whey by applying the nanofiltration unit (NF) reaches a dry matter content between 16% and 20%. The scheme in Fig. 5 shows the set-up of the nanofiltration step. Here, the retentate stream is recirculated to the feed vessel and the permeate stream is removed from the process until the feed solution has the desired dry matter content.

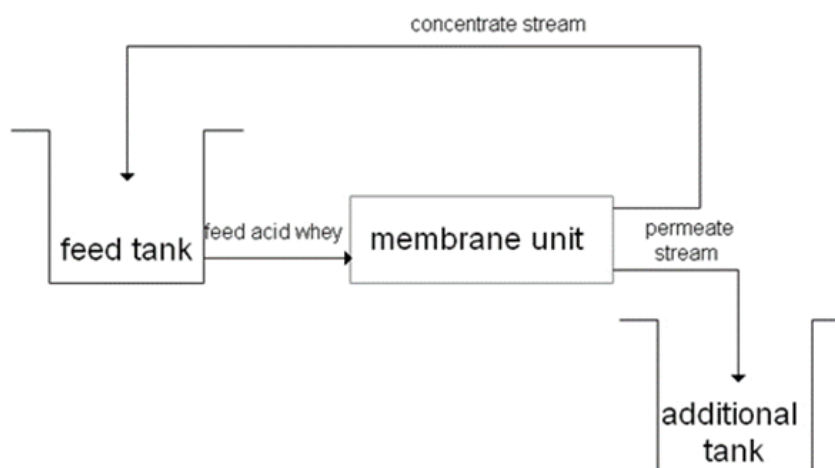


Fig. 5 Scheme of the pilot nanofiltration unit

3.1.2 ENM & NF for acid whey thickening in order to produce a substrate for biogas production

An alternative for the treatment train for the valorization of acid whey as biogas substrate or as soil conditioner as outlined in chapter 3.1.1 is the replacement of the centrifuge in this treatment train by an electrospun nanofibrous membrane (ENM). According to Tlili and Alkanhal (2019), the solid-liquor separation by using an ENM is a cost efficient alternative to a

centrifugation step. However, up to now, that technology was mainly applied in laboratory testing stages and still lacks its large-scale commercialization.

In Circular Agronomics, this technology will be investigated in detail aiming to show, that it is economically and ecologically rewarding.

Description of the treatment train or technology

The treatment train consists of two plants, the ENM and pilot-scale nanofiltration units. The acid whey inlet stream is treated by ENMs to reduce its fat content since its increased presence has negative effects on the membrane performance during nanofiltration. The acid whey with a low fat content is pumped into the pilot-scale nanofiltration unit equipped with a membrane of the type DOW Filmtec NF 3840/30-F. The feed solution results then in the permeate stream and the concentrated stream called retentate. The set-up of the nanofiltration unit is the same as the one in Fig. 5.

3.1 CE technologies for the recovery of N

For the production of a nitrogen fertilizer, usually a treatment train of at least two technologies is needed. In a first step, the nitrogen has to be recovered from the wastewater for example via ammonia stripping. Different stripping technologies exist such as air stripping, steam stripping, vacuum degasification and membrane stripping. In the second step, the ammonia is further processed into a nitrogen fertilizer. Therefore, absorption systems are applied. Those use acids or water to harvest the stripped ammonia either via spray systems (spraying liquid into gaseous phase) or via aeration systems (blowing gaseous phase into liquid).

Besides air stripping for the recovery of ammonia gas as already described in the BREF-document (2019), further stripping technologies exist such as membrane stripping and vacuum degasification. Those are compared in Tab. 4.

Tab. 4 Comparison of different stripping technologies such as vacuum, air and membrane stripping

Parameters	Units	Vacuum degasification	Air stripping	Membrane stripping
Source material	-	Sludge or sludge water	Sludge water	Sludge water
Pre-treatment (solids)	-	none	dewatering	dewatering & filtration
Total solid content (max)	mg TS/L	70,000	5,000	1,500
Pre-treatment (pH)	-	CO ₂ -stripping recommended to reduce NaOH demand		
NaOH (50%) consumption	kg/(kg N _{in})	6 (if CO ₂ is removed previously)		
Heat consumption	kWh _{th} /m ³	10 (heating from 30°C to 60 °C)		
Electricity consumption (NH ₃ + CO ₂ stripping)	kWh _{el} /m ³	2.0 + 0.6	1.6 + 0.6	2.4 + 0.6 (incl. filtration)
Literature	-	CIRC (PONDUS)	DEMOWARE D3.2	POWERSTEP 4.2/4.3

Energy consumption

The specific electricity consumption of the stripping technologies differs per technology. They range between 2.0 and 3.0 kWh/m³. The heat as well as sodium hydroxide consumption are similar. If CO₂ and/or carbonate is removed previously, the caustic soda demand depends mainly on the ammonium concentration and the related ammonia/ammonium buffer and not so much on the targeted pH value. The main distinction between the technologies is in their capability to manage solids within the stream. While membrane stripping requires extensive pre-filtration steps to reduce the total solids content to a minimum and protect the membrane, vacuum stripping allows to treat also sludge with a total solid content of up to 7%. Hence, as long as the total solids content does not exceed 7%, the energy consumption of the vacuum degasification is lower compared to the other stripping technologies, taking into account the energy consumption of the necessary dewatering step prior to the stripping process. According to DWA-M 366 (2011), that energy consumption ranges between 0.6 and 2 kWh/m³ depending on the dewatering technology. Hence, the minimum energy consumption for air and membrane stripping including an upstream dewatering step is assumed to be (2.2 + 0.6) kWh/m³ and (3.0 + 0.6) kWh/m³, respectively. That is slightly higher than (2.0 + 0.6) kWh/m³ for the vacuum degasification process without dewatering.

Regarding sorption systems, sulfuric acid is commonly used to harvest ammonium sulfate solution. The choice for sulfuric acid is due to its cheap price, its availability and high safety standards on the resulting product (e.g. ammonium sulfate compared to ammonium nitrate and the use of nitric acid). However, the monetary value of the obtained product is comparably low (75 €/t N), compared to 500-1000 €/t N for solid N fertilizers. Concentration, crystallization or drying of ammonium sulfate is difficult due to its high solubility and hygroscopic behavior.

Adsorption chemicals and related N products

An alternative system producing ammonium sulfate solution is realized by the BENAS plant in lower Saxony (Germany), using gypsum to harvest ammonia and carbon dioxide to produce calcite and ammonium sulfate solution. A system using nitric acid as sorption material is implemented in Oslo (Norway) and partly operated by YARA.

To achieve higher revenues in the future for recovered ammonium salts, solid salts should be recovered. Fig. 6 shows exemplary some relevant solubility equilibria for ammonium salts, whereby it shows that $\text{NH}_4\text{H}_2\text{PO}_4$ (MAP) or NH_4HCO_3 are salts which already form at low temperatures and hence, compared to the other salts are easier to recover.

While MAP is a commonly used phosphate fertilizer, the usage of NH_4HCO_3 is limited to China, where 8% of the N-fertilizer is applied in the form. A solid business case with MAP and the use of phosphoric acid is unlikely, due to the limited access and its high price. NH_4HCO_3 on the other hand, could be used as a potential intermediate for upcycling the included ammonia into different fertilizers. CO_2 and carbonic acid as scrubber acid, have the advantage that they are in most cases available and stripped prior to ammonia to reduce the buffer capacity of the liquor.

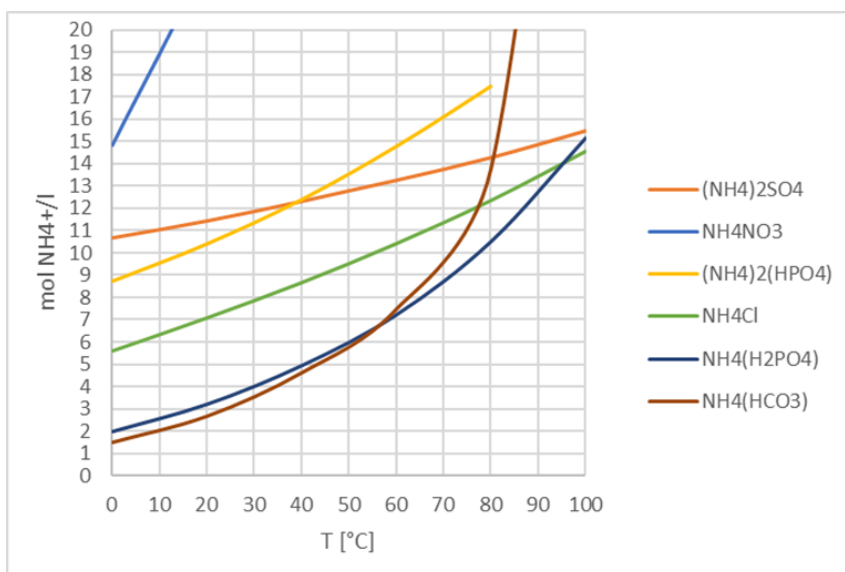


Fig. 6 Solubility of different ammonium salts (own figure according to <https://wissen.science-and-fun.de/tabellen-zur-chemie/loslichkeit-anorganischer-verbindungen-2/>)

3.2.1 Recovery of ammonia gas via vacuum degasification and gas scrubbing for ammonium sulfate production

In Circular Agronomics, different organic waste and wastewater streams from food and agricultural industry are investigated in terms of N recovery with vacuum degasification. Especially waste(water)streams with a high ammonium content such as digestates or manure are suited for this technology. The vacuum degasification unit is derived from a methane vacuum degassing unit for digested sewage sludge (TRL 9). In Circular Agronomics, this technology is further developed to an ammonia vacuum degassing unit (TRL 5) with a subsequent gas scrubber for the production of ammonium sulfate (TRL 9). Therefore, a pilot plant with a flow rate of 50 L/h is designed and constructed.

Description of the treatment train

The pilot plant consists mainly of two units, the vacuum degasifier and the gas scrubber (Fig. 7). The substrate is filled in the substrate tank, which is connected to a cutter in order to shred solids contained in the substrate. Substrates containing a total solid content of up to 7% can be processed in the pilot plant. The substrate stream can be circulated through the cutter as long as necessary until the solids are small enough for entering the heat exchanger and the degasifier. Here, if the pH is at neutral conditions, CO_2 can be removed via vacuum degasification. Furthermore, after the pH adjustment to alkaline conditions via NaOH and still at an elevated temperature, ammonium reacts to ammonia and can be degassed as well.

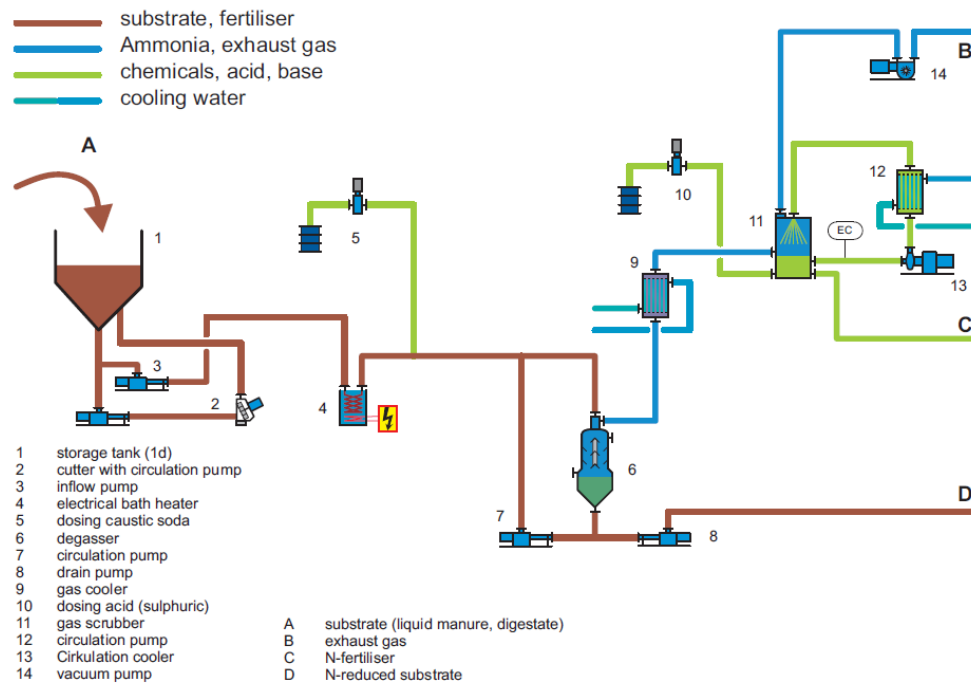


Fig. 7 Process scheme for vacuum degasification of ammonia gas with its subsequent scrubbing in sulfuric acid

The crucial process parameters such as pH, temperature and pressure can be varied in different ranges. In general, the pH will be adjusted between 8 and 10, the temperature can be varied between 20 °C and 70 °C and the pressure can be adjusted between 100 mbar and 900 mbar (indicated as absolute pressure). In the subsequent gas scrubber, sulfuric acid is circulated in order to react with the ammonia gas to ammonium sulfate. In agriculture, ammonium sulfate is also called ammonium sulfate solution. This is a typical mineral nitrogen fertilizer. After ammonia removal via degasification from the material, the so called “nitrogen depleted” material is ready to be applied as a soil conditioner.

3.2 CE technologies for the recovery of P

For the recovery of phosphorus and its reuse for fertilizing, more than 70 different processes and treatment trains already exist especially for the recovery from municipal wastewater and sludge (Roskosch et al. 2018, Kraus et al. 2019). In Germany for example, there are already six large scale plants in operation (Kleyböcker et al. 2019). In addition, the BREF-document (2019) considers struvite recovering technologies as BAT as already outlined in chapter 2.3.3.

Depending on the pH in the P recovery process, the crystalline form of the calcium phosphate compound varies. At a pH between 5 and 6, brushite dominates, while between pH 6 and pH 7, octacalcium phosphate is mostly observed and at pH 7 and above, hydroxyapatite forms (Seckler et al. 1996, Montastruc et al. 2003). As already described in the BREF-document, for the dairy industry, the precipitation of hydroxyapatite after lime dosing is listed as BAT. In this chapter, the P-recovery technologies being developed and investigated in Circular Agronomics will be described in detail.

3.3.1 Enzymatic enhanced phosphate release with subsequent struvite production

Some major food and feed substrates do contain phosphorus as phytic acid, a multiple P containing cycle molecular structure. One of the most important feed/food additives in this regard is soya bean. Phytic acid has a known limited degradation during anaerobic wastewater treatment. This renders phosphate (PO_4) recovery by means of struvite formation difficult. In the subsequent aerobic stage the degradation of phytic acid occurs, releasing the P as PO_4 and requiring significant dosing of ferric or aluminum salts to attain the final P discharge levels for the effluent. If phytic acid conversion into PO_4 could be enhanced during anaerobic processing, this would result in a wastewater composition suitable for struvite formation. In addition, analysis of compounds show that in this particular case PO_4 would be the limiting parameter rather than magnesium. The selected approach to reach higher PO_4 levels is to increase the phytase enzymatic

activity by either added commercial available phytase enzymes or by cultivating *in vivo* high phytase producing fungi or yeasts.

Description of the treatment train

The pilot unit has a multiple purpose and can be run in different scenarios at an average flow of 1 m³/h. Fig. 8 gives an overview of the general wastewater and solid waste process flow and how they are processed at this stage in the full scale soya bean processing plant. The possible interaction with added phytase enzymes to improve the conversion of bound-P into to recoverable PO₄ are indicated as option 1 and option 2.

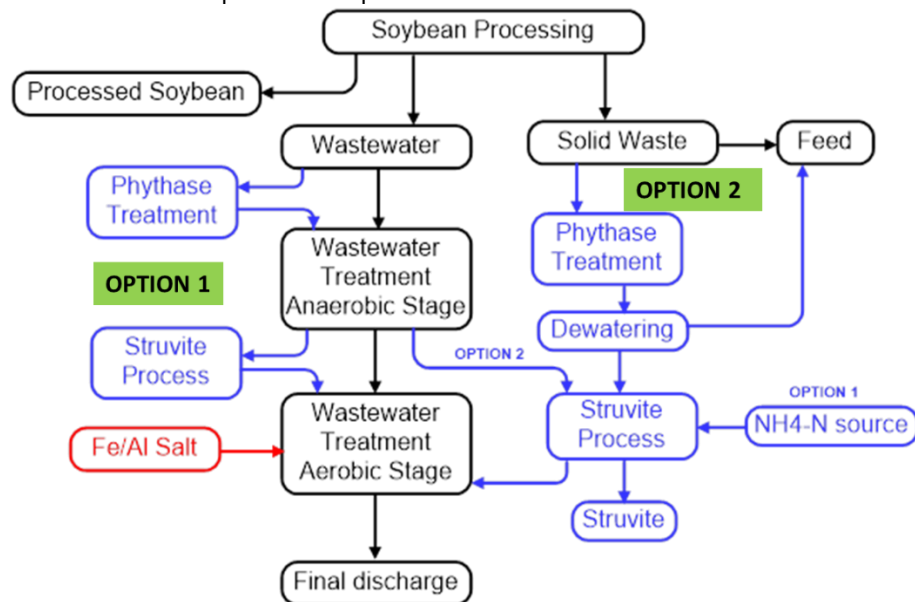


Fig. 8 Main stream processing of wastewater and solids wastes.

The major gain can be achieved on the main wastewater flow. The normal processing is a straight forward anaerobic treatment after buffering and equalization. The wastewater is at 40-50 °C and pH of 4.5-5.5. This requires some cooling by active heat exchange. Given the low pH and elevated temperatures these conditions do favor selected growth of fungi/yeasts and are in line with optimal process conditions for a number of commercial available phytase enzymes. One should take note that this low pH and lack of free ammonium does prevent the struvite formation at this stage. The latter is also the case for possible K-struvite formation, the pH at this stage is too low.

In the adapted flow the wastewater is pre-treated with phytase enzymes. These enzymes are a bulk commodity already largely used as feed additive to improve digestibility. By adding the selected phytase enzyme(s) the phytic acid is hydrolyzed prior to entering the anaerobic stage. Since there is no or only little PO₄ uptake of the liberated phosphate, that will be incorporated into new anaerobic cell biomass (there is only limited sludge growth under anaerobic conditions), most of the produced PO₄ will remain in solution and be present in the anaerobic stage effluent. In addition, the anaerobic treatment will also convert organic compounds liberating the contained nitrogen as ammonium (major constituent of struvite) as well as increase the pH. Under these conditions, high levels of PO₄, NH₄ and an elevated pH are reached and the formation of struvite can be induced. This results in phosphorus removal/recovery between the anaerobic and aerobic stage of the wastewater treatment. An alternative approach would be instead of adding commercial phytase enzymes, trying to induce *in vivo* phytase production in the buffer prior to the anaerobic stage. Since there are no indications that *in vivo* phytase production occurs at this stage a separate cultivation reactor, feeding continuously phytase active fungi/yeasts in the buffer, might be needed to work according to this approach.

Finally a last possibility (option 2) is to investigate, if phytase treatment can release additional PO₄ contained in the separated solids.

Fig. 9 shows, how the different options 1 and 2 can be applied, and their locations within the existing treatment processes. The enzyme aqueous solution can be prepared separately starting from purchased enzymes as a powder and dosed at a given ratio versus the incoming flow into a separate hydrolysis reactor. The *in vivo* phytase activity can be obtained by cultivating selected fungi/yeasts strains in a separate correctly conditioned reactor (pH: 3.5 – 4.5 and temperature: 40-45 °C). The bypass of incoming wastewater can be used as feed source to induce phytase production. The *in vivo* reactor

content can either be introduced as such in the phytic acid hydrolysis reactor or after the separation of the active biomass, the processed liquor can be used for that. These additional treatments on the wastewater flow are aimed at increasing the available PO_4 which at an elevated pH and with liberated ammonia can be converted into NH_4 -struvite. The latter can be done by either a pH increase (given the high level of magnesium (Mg) already present in the wastewater) or by adding extra Mg to achieve higher recovery rates. Processing the solids is the second option. A blended mixture of these solids and phytase enzymes (commercial product) can be incubated to release additional PO_4 . An extra dewatering phase would thus generate extra PO_4 for struvite formation.

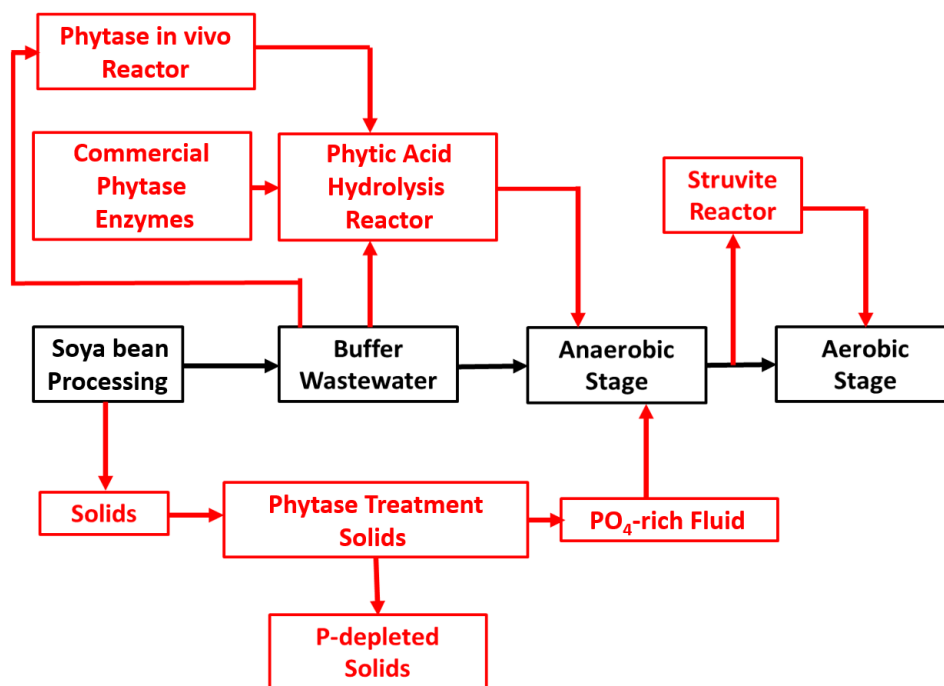


Fig. 9 Black flow is the current treatment/ red flow is the proposed possible phytase treatments

3.3.2 Enzymatic enhanced phosphate release with subsequent K-struvite production

Phosphorus related issues have been described already in 3.3.1. *Enzymatic enhanced phosphate release with subsequent struvite production*. Since phosphorus is a limited resource and essential for food/feed production, a fertilizer reuse will be mandatory to ensure food safety in general for future generations. Besides nitrogen, the other key fertilizer compound is potassium. Also, potassium is a mined resource with limited supply. Potassium is as opposed to N and P not a major constituent of biomass, but it is a key element in terms of metabolic activity and ensures good cation/anion equilibria and transmembrane transport mechanisms. The latter is inherently coupled with the high water solubility of K. This high water solubility is an extra challenge, when focusing on K-recovery since the first step in any type of recovery processes is most likely an extraction process in order to obtain a concentrated flow out of a diluted aqueous flow. A compound with low water solubility renders the latter feasible. One of the few K-salts with low water solubility is K-struvite (the analogue to main stream NH_4 -struvite). The general rule is that as long as NH_4 is present NH_4 -struvite will be produced, only when NH_4 is depleted and if at that moment still PO_4 is available, K-struvite will be formed. In addition, K-struvite requires a higher pH compared to NH_4 -struvite under similar conditions.

Description of the treatment train or technology

For the production of K-struvite, NH_4 needs to be depleted. Thus, K-struvite formation is only possible after the aerobic stage. The wastewater has a potassium concentration between 1000 and 1500 ppm that is also observed in the aerobic stage effluent. A part of the dissolved Mg in the effluent from the anaerobic stage precipitates in the aerobic stage, but the other part will still be dissolved and available in the aerobic stage effluent. The key parameters to control the precipitation process are the pH and, if needed, additional dosages of Mg. Fig. 10 shows the implementation in an existing plant. The technology train will require a struvite crystallization reactor but also a change in the overall P management. If the goal is K-struvite formation after the aerobic stage, no addition of Fe/Al in the aerobic stage is allowed in order to maximize the

D3.1. Classification of food waste and wastewater streams

PO₄ throughput towards the K-struvite unit. Also the final PO₄ obtained after K-struvite formation will be in order of 20-30 ppm PO₄-P. Hence, there will be the need for an additional tertiary P-removal after the K-struvite production in order to attain the required final discharge levels. In most cases, a type of tertiary P-removal is already implemented. The K-struvite process is applied on the entire flow of the wastewater.

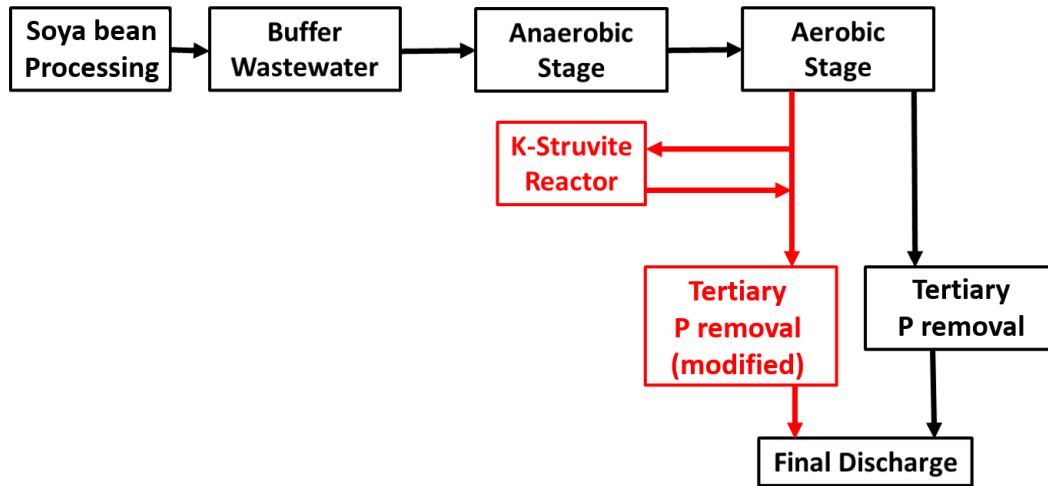


Fig. 10 Implementation of the K-struvite process in an existing plant

3 Classification of waste(water) streams in terms of the application of the technologies

Waste and wastewater streams from the food, drink and milk industry vary over a wide range regarding their contents of dry matter (DM), total suspended solids (TSS), organic carbon indicated as chemical oxygen demand (COD), nitrogen (N), phosphorus (P) and potassium (K). In this chapter, based on those parameters, 26 waste streams and 20 wastewater streams (Tab. 5) are classified and evaluated for their suitability for the novel technologies being developed in Circular Agronomics.

Tab. 5 List of the characterized waste and wastewater streams: same colored letters indicate a similar origin of the streams

Waste streams		Wastewater streams from	
1	Wheat mash from distillery	1	Distillery
2	Cereal mash from distillery	2	Slaughterhouses
3	Potato mash from distillery	3	Rendering production
4	Rumen content (untreated) from slaughterhouses	4	Vegetable processing
5	Flotation sludge from slaughterhouses	5	Dairy
6	Meat and bone meal from slaughterhouses	6	Breweries
7	Rumen content (pressed) from slaughterhouses	7	Fruits processing
8	Blood meal from rendering	8	Wineries
9	Stomach content of pigs from slaughterhouses	9	Potato processing
10	Vegetable wastes from vegetable processing	10	Potato starch production
11	Market wastes from vegetables	11	Wheat starch production
12	Whey (normal & thickend) from dairies	12	Corn starch production
13	Beer spent from breweries	13	Sugar production
14	Spent diatomite (beer) from breweries	14	Soft drinks production
15	Spent hops (dried) from breweries	15	Gummi candy production
16	Leftovers canteen kitchen	16	Gelatine production
17	Spent apples from juice production	17	Pectin production
18	Spent fruits from juice production	18	Fisheries
19	Vine spent from wineries	19	Olive oil mills
20	Vinasse from sugar production	20	Soya bean processing
21	Molasses from sugar production		
22	Dry bread from bakeries		
24	Colza extraction shred from oil mills		
25	Oilseed residuals from oil mills		
26	Solid waste from soya bean processing		

Therefore, most values and ranges for the relevant parameters were obtained from literature. Data for the waste streams refer to Deublein et al. (2008), Rosenwinkel et al. (2015), to own data for solid waste from soya bean processing (Moermann 2020) and own data from Holba et al. (2020) for whey. For the wastewater related data, those were collected from Bischofsberger et al. (2005), Puchlik et al. (2017), Abdel-Fatah et al. (2015), Eremektar et al. (2002), Övez et al. (2001), Poddar et al. (2017), Waldron et al. (2007), Rosenwinkel et al. (2015), Laginestra et al. (2016), Silvano et al. (2018), Arturi et al. (2019), Hung et al. (2006), Auterska et al. (2006), Doble et al. (2005), Wathugala et al. (1987), Ching and Redzwan (2017) and for wastewater from soya bean processing own data were used from Moermann (2020).

4.1 Waste(water) streams with high C content

In general, waste and wastewater streams can be distinguished due to their contents of DM and TSS. While waste streams contain a relatively high dry matter contents ranging from 3% DM up to 98% (Fig. 11), wastewater streams contain much lower DM contents usually below 7% DM. However, due to a convention, the typical parameter for wastewater is usually the TSS content and not the DM content. That ranges for wastewater from the food, drink and milk industry typically from 8 mg/L to 57,000 mg/L (Fig. 12).

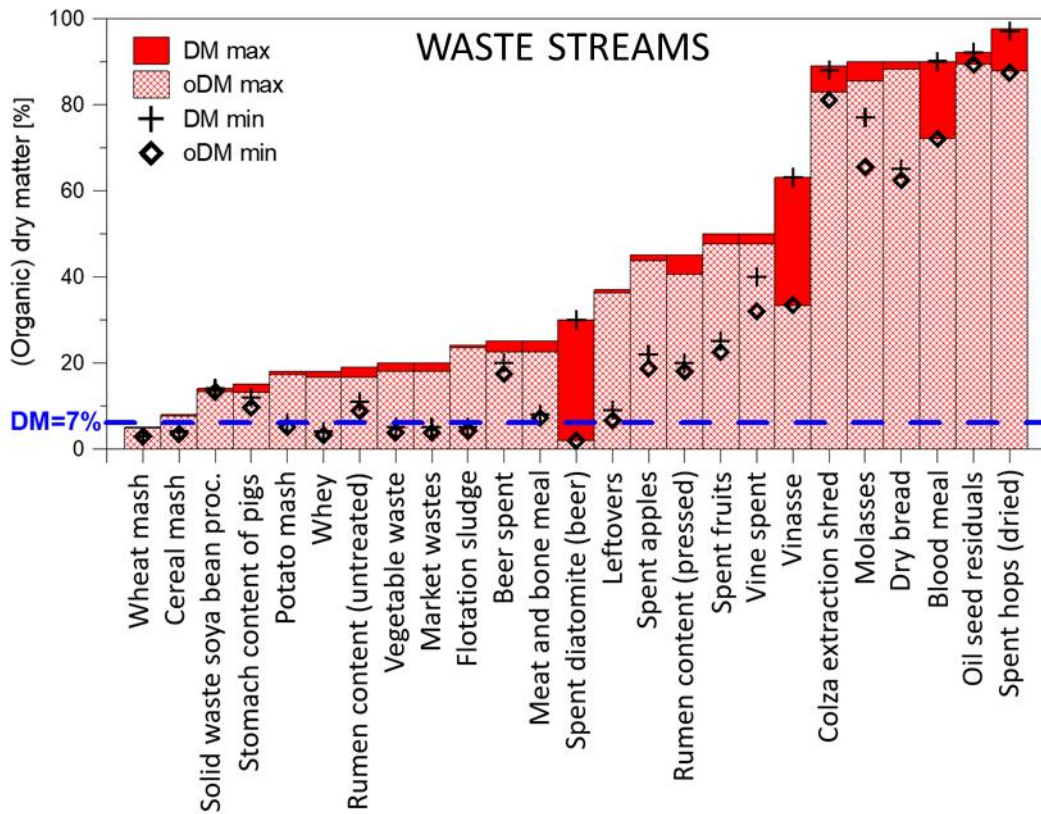


Fig. 11 25 waste streams characterized by their dry matter (DM) content and their organic dry matter (oDM) content: ranging from 3% up to 98%.

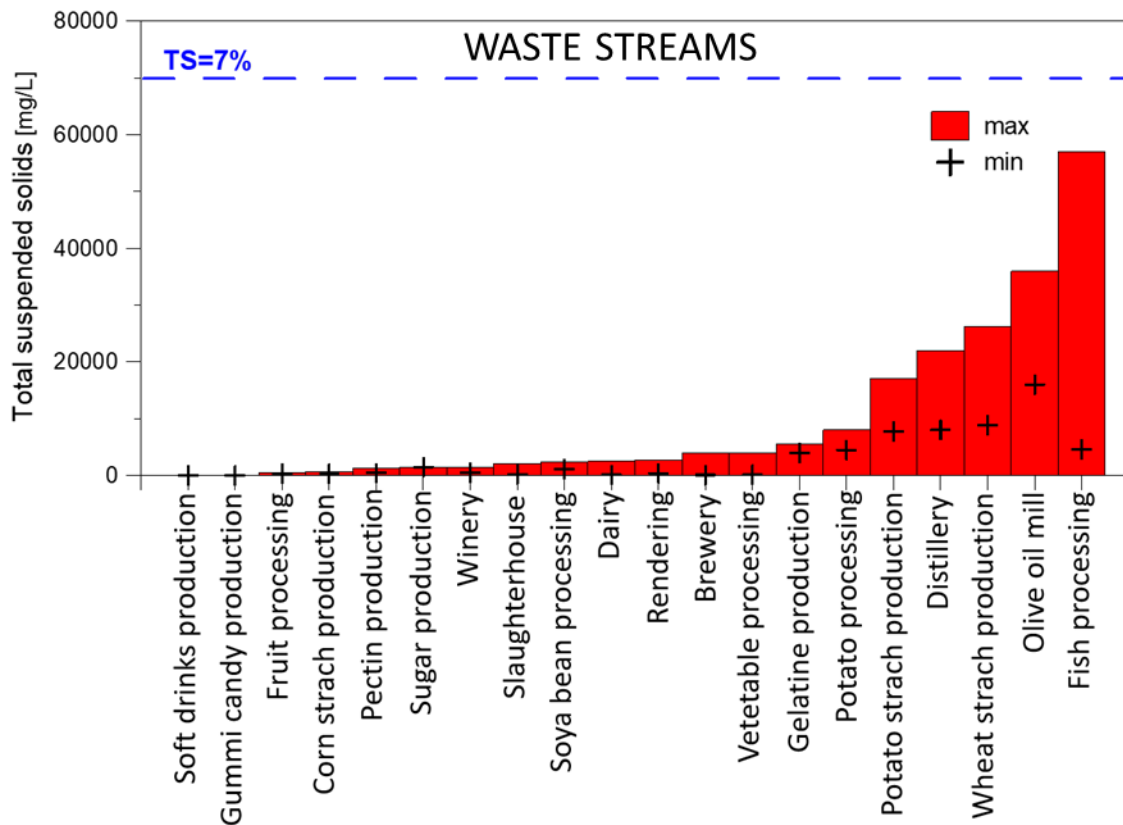


Fig. 12 20 wastewater streams characterized by their total suspended solids content: ranging from 8 mg/L to 57,000 mg/L

D3.1. Classification of food waste and wastewater streams

As shown in Fig. 11, the organic dry matter (oDM) content is also usually very high in those waste streams ranging from 80% of DM to 98% of DM, besides a few exceptions such as for spent diatomites from beer production and for vinasse. Thus, due to the high contents of organic matter those waste streams are well suited as (co-)substrates for biogas production. Fig. 13 shows the expected methane yields from those waste streams. They range between 0.2 m³ CH₄/(kg oDM) and 0.8 m³ CH₄/(kg oDM). The methane yields indicate, that the waste streams contain a high fraction of volatile organic matter, which can be easily digested in order to produce biogas.

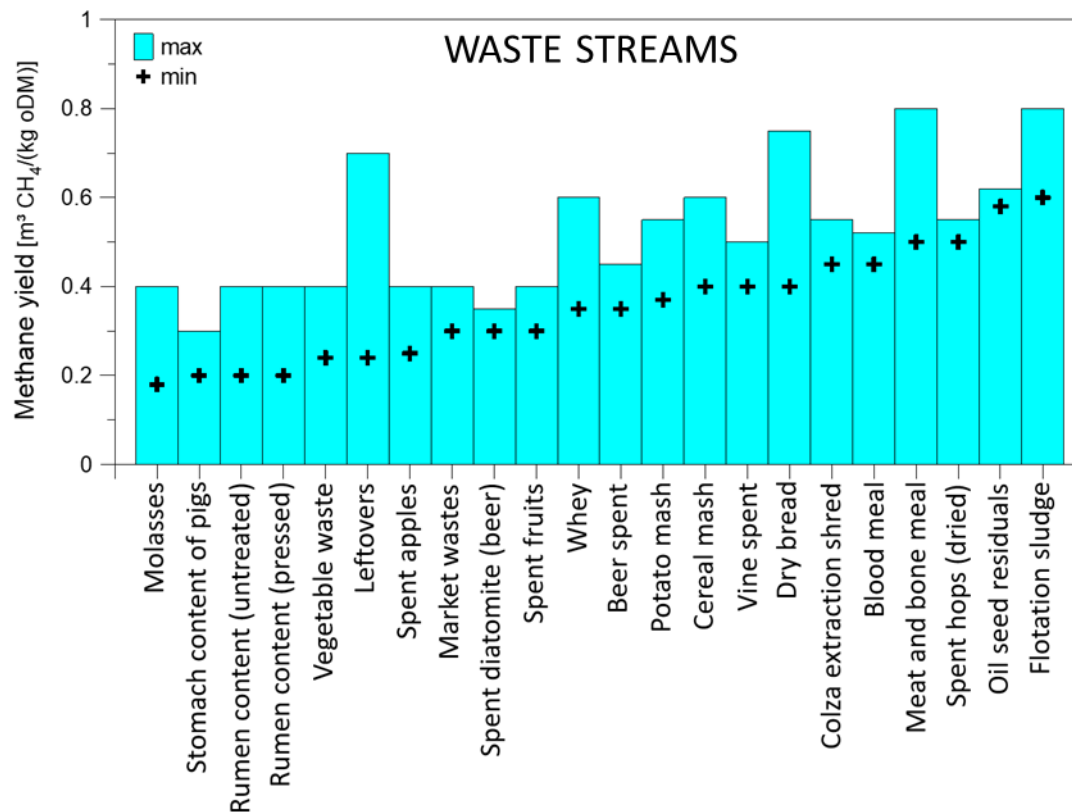


Fig. 13 Expected methane yields for different waste streams from the food, drink and milk industry.

Regarding the wastewater streams resulting from the food, drink and milk industry, their concentrations of the chemical oxygen demand (COD) range from 200 mg/L up to 150 g/L (Fig. 14). According to Bischofsberger et al. 2005, Waldron 2007 and Rosenwinkel et al. 2015, all wastewater streams are all very suitable for anaerobic digestion since the biodegradability of their organic contents is high and ranges between 60% to 99%. Especially for wastewaters from fish processing, olive oil mills, wheat starch production, distilleries and potato processing, their biodegradabilities are between 70% and 95% (Bischofsberger et al. 2005, Dhanke et al. 2020, Krzywonos et al. 2009). Depending on their pH, a pH adjustment might be necessary prior to anaerobic digestion. However, as it will be presented in the next chapter, the pH varies widely depending on the certain food processing processes applied by the industries (see also Fig. 18). Thus, for the concrete treatment, the wastewater should be characterized first for designing the right treatment train.

Especially for wastewaters with COD concentrations of 1000 mg/L and above, anaerobic digestion is well suited for energy recovery (Grady et al. 1999, Möbius 2010). Due to the usually dissolved form of the COD, the carbon is very well available for microbial processes and thus, for biogas formation. Fig. 14 shows, that the maximum COD concentration of every wastewater reaches 1000 mg/L and more. However, the minimum ranges for wastewaters from soft drinks production, fruit processing, sugar production, vegetable processing and slaughterhouse wastewater are far below 1000 mg/L and thus, for the concrete application, the actual COD content should be estimated or determined, if the implementation of an anaerobic treatment is considered. The highest potential for energy recovery in the form of biogas have the wastewaters from fish processing, olive oil mills, wheat starch production, from distilleries, from potato starch production and from gummi candy production due to their high COD concentrations.

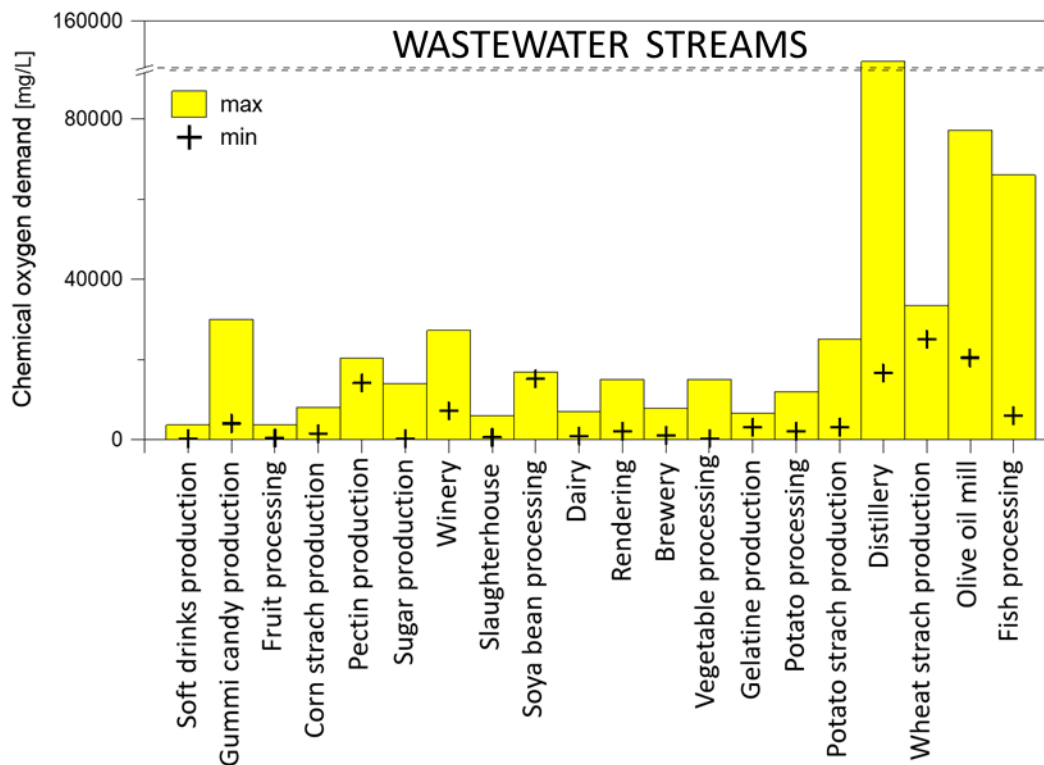


Fig. 14 Chemical oxygen demand concentrations of 20 different wastewater streams resulting from the food, drink and milk industry ranging from 200 mg/L to 150 g/L.

A direct application on the fields as soil conditioner for example is widely not recommended, because the volatile organic compounds can also be easily degraded aerobically on the field and be emitted as CO₂ into the air. Simultaneously, the carbon is lost for the soil improvement and/or for carbon sequestration. Furthermore, due to the aerobic degradation, ammonium and nitrate might be formed. If the soil pH is alkaline, the ammonium reacts to ammonia and degasses into the air. In addition, due to the aerobic conditions, nitrate might be formed. Nitrate is highly water soluble and prone to leaching (Haag and Kaupenjohann 2000): Thus, if it is not denitrified due to a lack in available organic carbon or due to an aerobic milieu, it is easily emitted into the groundwater. Moreover, if the nitrification process of the ammonium is not completed, nitrous oxide may occur, having the impact of 298 CO₂ equivalents on the greenhouse effect.

However, a direct application on the fields without high emissions might be possible, if the pH of the soil is below 6.5 and only during the vegetation period, when the demand of the plants for ammonium or nitrate is high enough. It should be noted, that this is only a very specific case, which occurs a very limited time per year.

Conclusion for CIRCULAR ECONOMY regarding C rich waste and wastewater streams: anaerobic digestion is recommended

In order to avoid NO₃, CO₂, NH₃ and/or N₂O emissions due to an agricultural application on the fields, prior to that application, anaerobic digestion is widely recommended for C-rich wastes and wastewater streams in order to recover energy in the form of biogas. Here, the joint treatment of waste and wastewaters resulting from the same industry may lead to the advantage, that the DM content of the waste will be diluted by the wastewater resulting in better conditions for e.g. mixing and the pH which is for anaerobic digestion required to be in a neutral milieu. This should be investigated for each site, because the characteristics of the waste and wastewaters for each category vary widely as shown in the all the figures. In addition, during digestion, organic nitrogen is anaerobically degraded to ammonium and organically bound phosphorus can be also partly released as phosphate. Thus, the resulting digestate is very suitable for nutrient recovery technologies as described in chapter 3. After the nutrient recovery and hence, the nutrient depletion of the digestate, the remaining material is still suitable as soil conditioner with the advantage to be mainly depleted from compounds that might lead to emissions. However, especially for substrates with a high nitrogen content, an ammonium or ammonia inhibition of methanogenic archaea might occur starting from concentrations between 1,500 mg NH₄/L and 10,000 mg NH₄/L as well as 80 mg NH₃/L (Deublein et al. 2008). In order to avoid this, ammonia recovery technologies can be implemented in a side stream in order to decrease the ammonia concentrations in the digester content.

4.2 Waste(water) streams with high N content

The nitrogen content of the considered waste streams ranges from 1.3% DM for solid waste from soya bean processing up to 13% DM for potato mach (Fig. 15). Considering the DM content of each waste stream, total nitrogen concentration levels between 540 mg/L for leftovers and 108 g/L for blood meal are reported (Fig. 16).

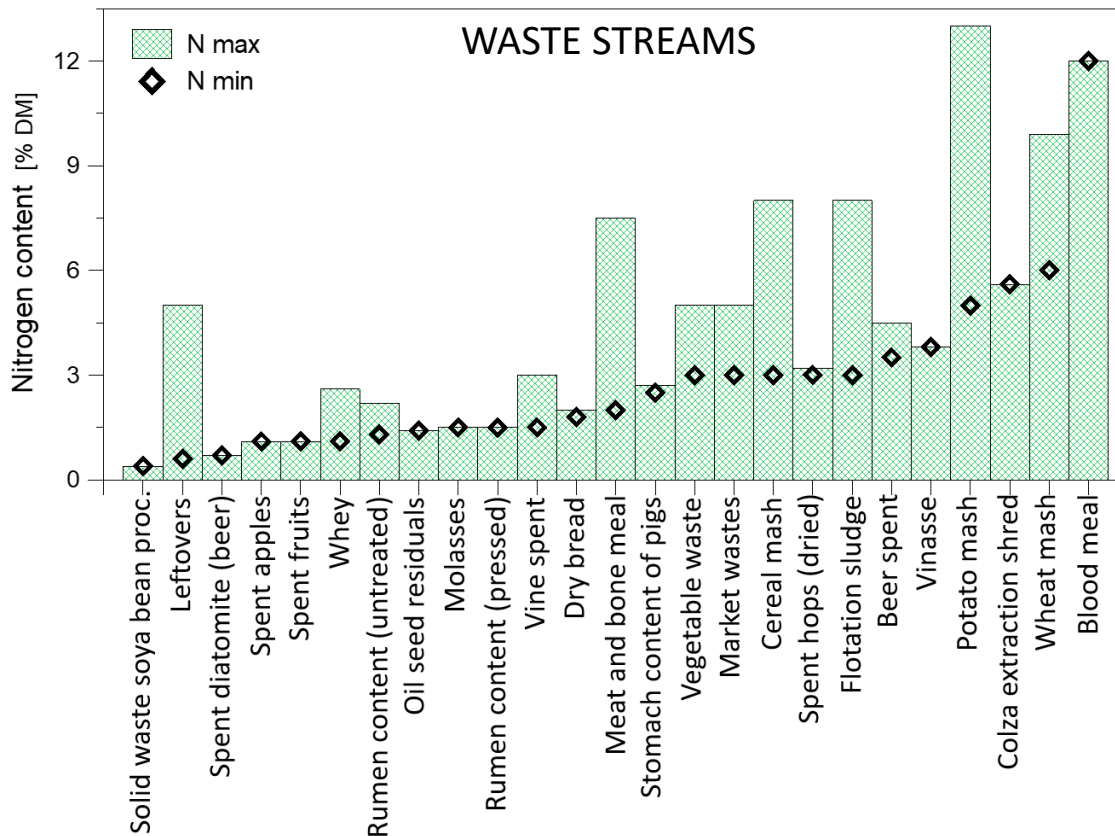


Fig. 15 Total nitrogen contents indicated as percentage of their DM content for 25 different waste streams

For some waste and wastewater streams, the ratio of ammonium nitrogen to the total nitrogen can be quite low as for gelatin and soya bean processing wastewater with 0.01% (Arturi et al. 2019, Rosenwinkel et al. 2015, Moermann 2020) or it is below 50% as for fishery wastewater with 17%, winery wastewater with 26% and slaughterhouse wastewater with 43% (Waldron 2007). According to Wilken et al. 2019, anaerobic degradation processes convert organic bound nitrogen into ammonium. In doing so, depending on the substrate type usually between 50 and 80% of the total nitrogen are converted to ammonium. Hence, assuming a conversion rate of 50% and the fact, that ammonium concentrations between 1,500 mg NH₄/L and 10,000 mg NH₄/L inhibit methanogenic archaea (Deublein et al. 2008), in Fig. 16, limits were defined for a possible inhibition and for a likely inhibition of the methanogens. It should be noted, that this would be only the case, if the substrates are digested without any further substrates and/or the co-substrates contain similar high nitrogen concentrations and thus, the ammonium concentration won't be diluted. Furthermore, if the pH of the digester content increases to a slightly alkaline milieu, the ammonium reacts to ammonia and an ammonia inhibition might occur.

Thus, based on those assumptions, first, the digestion process is crucial in order to generate enough ammonium and/or ammonia for its recovery. This intermediates are a prerequisite in order to apply the stripping technologies as suggested in Chapter 3. However, depending on the total nitrogen contained in the waste or wastewater stream, the implementation of the stripping technology is recommended either in a side stream of the digestion process, in order to avoid an inhibition of the methanogens, or in the main stream after the digestion process. The main stream implementation can be applied, if the concentration of a nitrogen rich waste(water) stream will be diluted due to other co-substrates or because the concentration is below the threshold for the inhibition.

In general, blood meal, wheat mash, potato mash, flotation sludge, cereal mash, meat & bone meal and leftovers have the highest share of nitrogen related to their DM content ranging between 5% and 13% of their DM. However, if a dewatering

D3.1. Classification of food waste and wastewater streams

treatment shall be avoided, it is necessary to consider not only the nitrogen share of the DM, but the actual concentration in the waste stream resulting from the industry. Therefore, in Fig. 16, the total nitrogen concentration in the waste streams are presented.

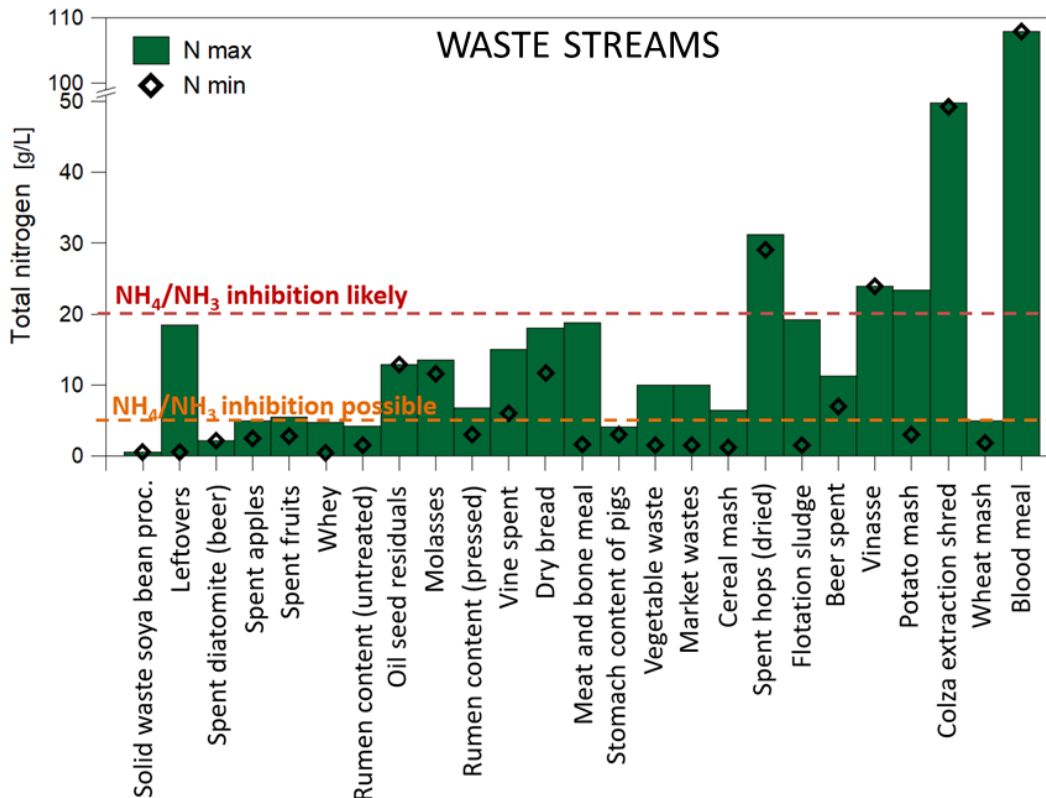


Fig. 16 Total nitrogen concentrations for the same waste streams as shown in Fig. 15 and in the same sequence. Thresholds for a possible and a likely inhibition of methanogenic archaea during anaerobic digestion were estimated according to Deublein et al. 2008 and Wilken et al 2019.

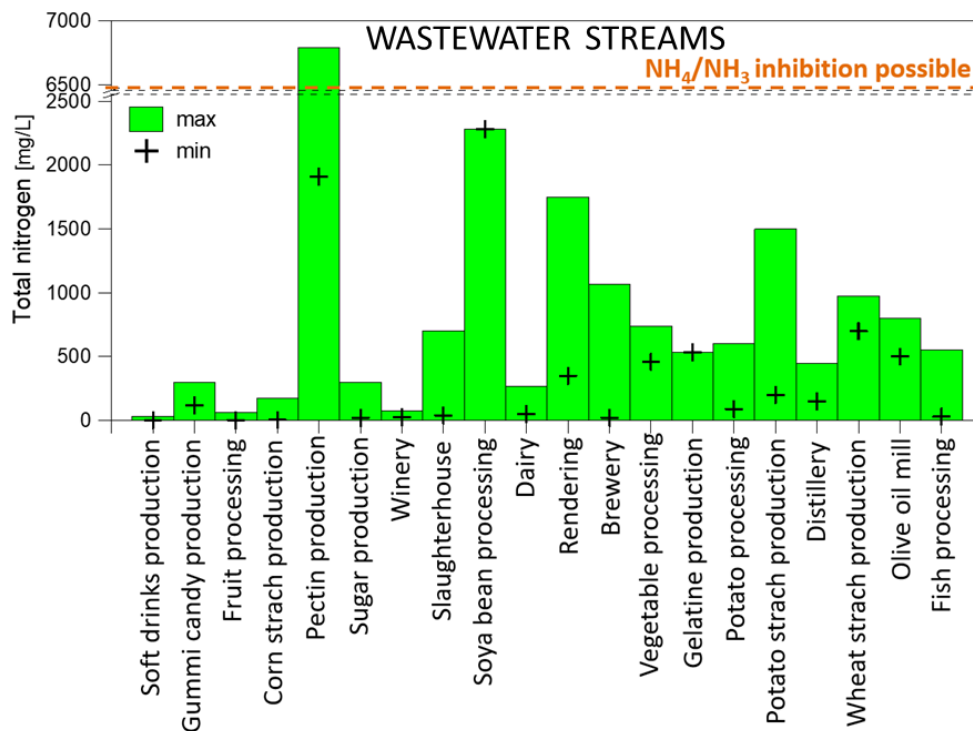


Fig. 17 Total nitrogen concentrations for the wastewater streams: threshold for a possible inhibition of methanogenic archaea during anaerobic digestion was estimated according to Deublein et al. 2008 and Wilken et al 2019.

According to Fig. 16, especially for blood meal, colza extraction shred, spent hops, vinasse and potato mash, the implementation of a stripping technology in a side stream for example in the re-circulation stream of a digester might be considered. In terms of the wastewater streams, Fig. 17 shows that most nitrogen concentrations are below the critical threshold. However, especially for wastewater from pectin production, the necessity of implementing it as a side stream should be investigated. Due to a high variety of the different wastewater streams in the pH as shown in Fig. 18, anaerobic digestion as a pretreatment can especially for the acidic substrates raise the pH due to the degradation of organic acids. Thus, in terms of the stripping process, the demand of chemicals such as sodium hydroxide for increasing the pH to 9 can be diminished.

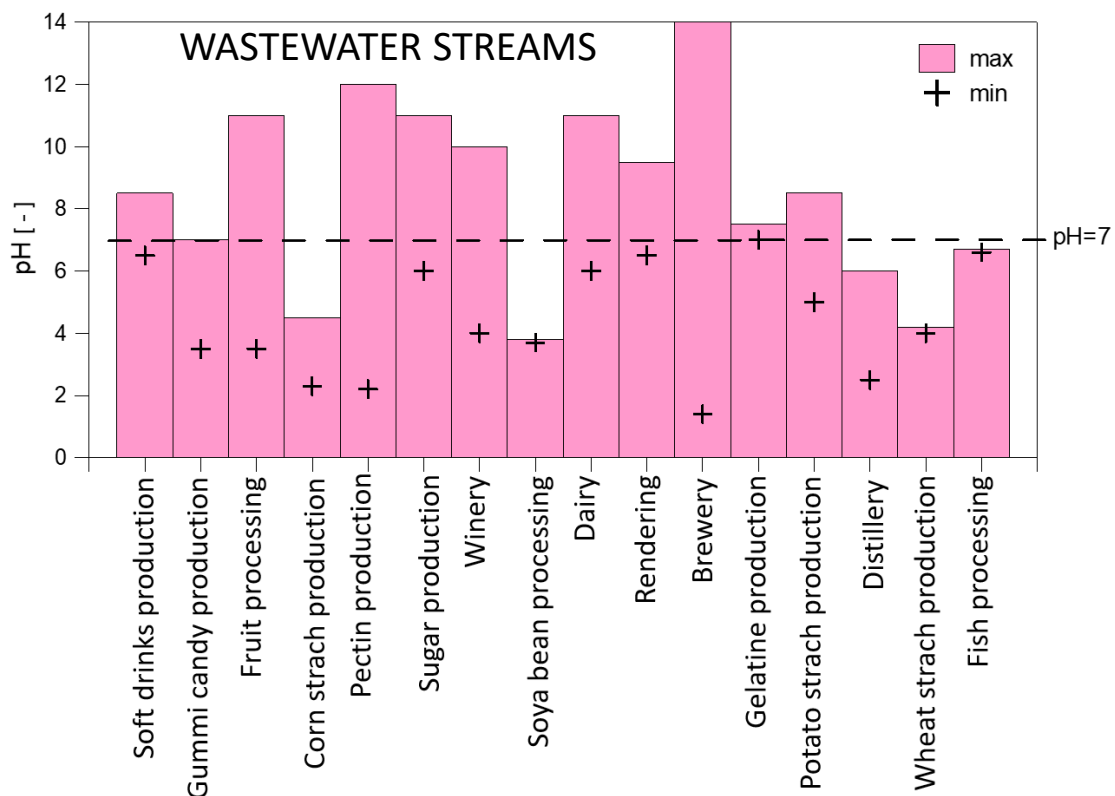


Fig. 18 pH ranges for different wastewaters from the food, drink and milk industry: depending on the certain case, the pH can vary widely from very acidic to very alkaline

Conclusion for CIRCULAR ECONOMY regarding N rich (waste)water streams: Anaerobic digestion and N depletion prior to the application as soil conditioner on the fields is recommended

As already concluded in 4.1, before waste(water) streams are applied on the agricultural fields, anaerobic digestion is widely recommended with a subsequent removal of nitrogen in order to avoid undesired CO₂, N₂O or nitrate emissions. Furthermore, if substrates with very high nitrogen contents shall be digested such as blood meal, colza extraction shred, spent hops, vinasse and potato mash, the ammonium is suggested to be removed in a side stream in order to decrease the ammonium concentration in the digester. Thus, a likely process failure due to an ammonium and/or ammonia inhibition of the methanogenic microorganisms in the digester can be avoided. Therefore, the implementation of a nitrogen removing technology such as the vacuum degassification technology is suggested since the technology maintains anaerobic conditions of the fluid. For a suitable technological concept see chapter 5.1.

4.3 Waste(water) streams with high P content and K content

In terms of the waste streams, two parameters, the phosphorus content indicated as percentage of the DM content and the phosphorus concentration indicated as mass per volume are considered. As shown in Fig. 19, the waste streams range between 0.1% DM and 2.6% DM.

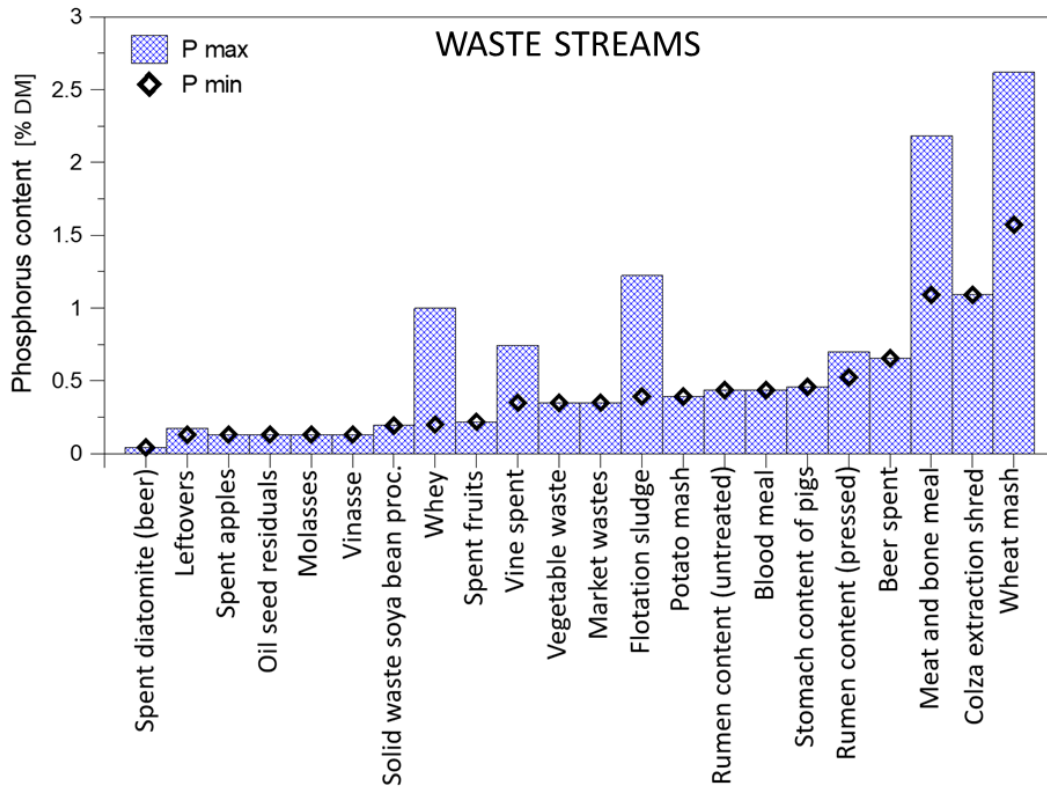


Fig. 19 Total phosphorus contents of different waste streams indicated as percentage of their DM content

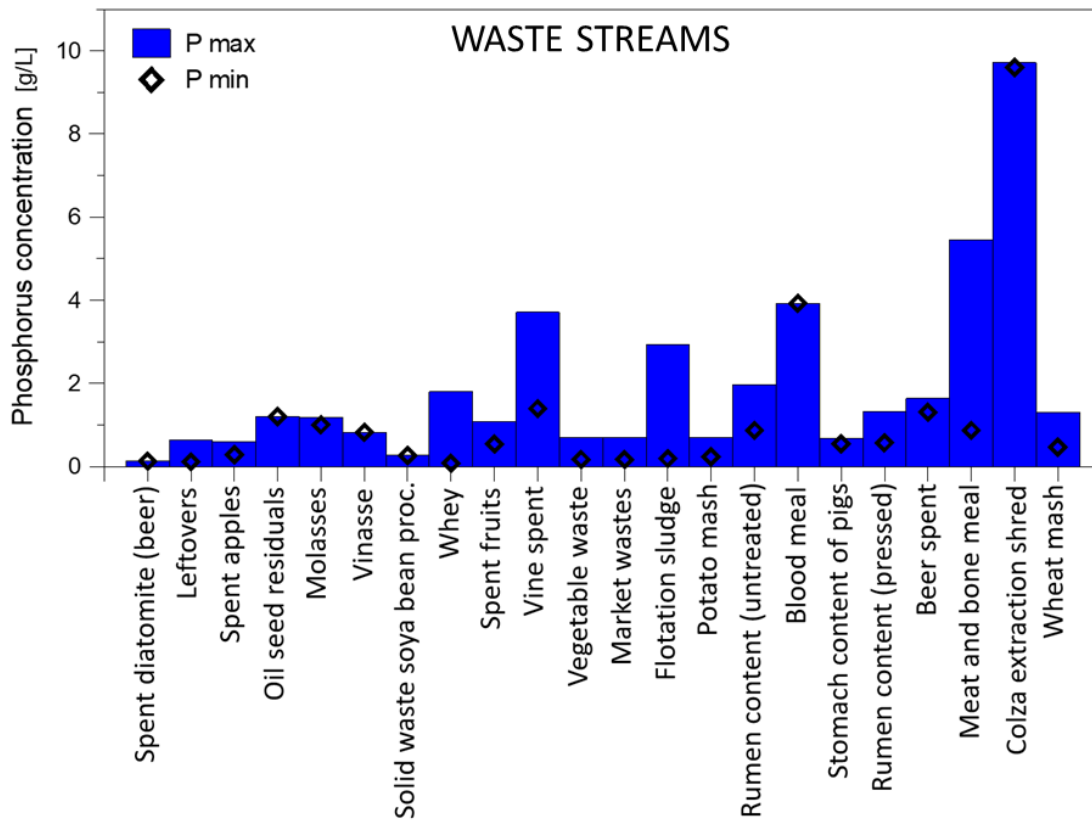


Fig. 20 Total phosphorus P concentrations of different waste streams

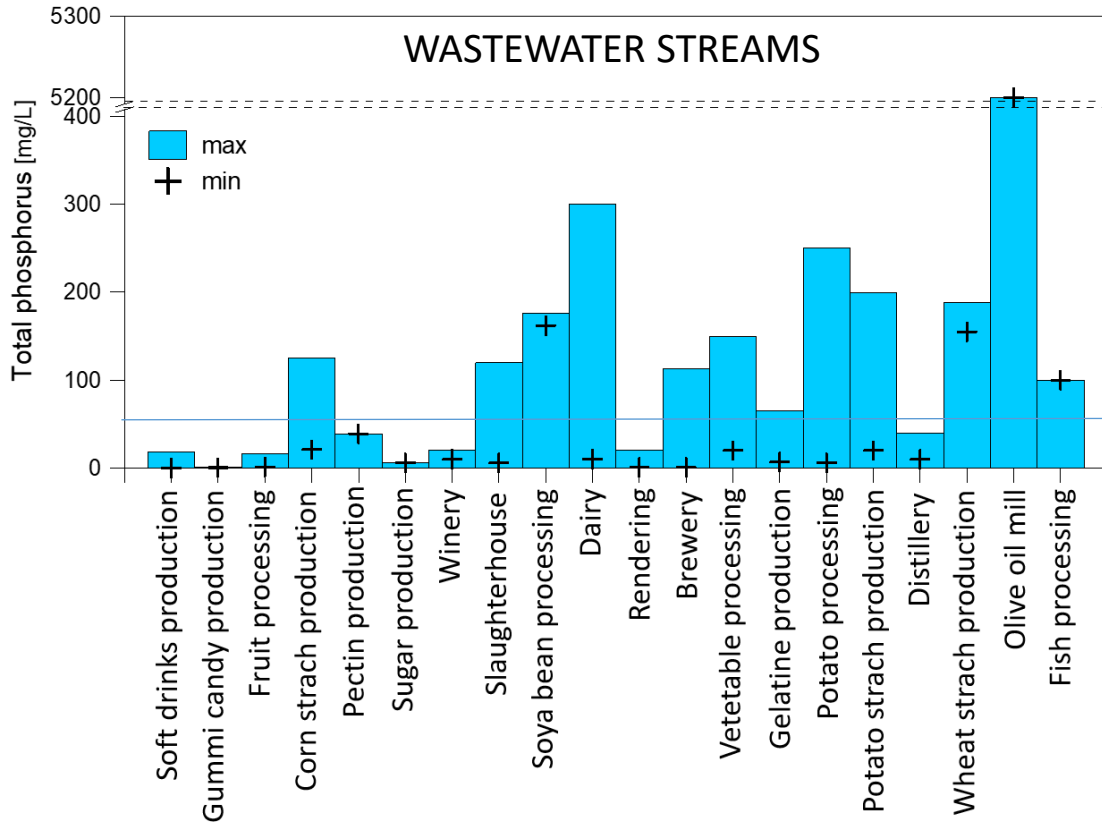


Fig. 21 Total phosphorus concentrations for different wastewater streams

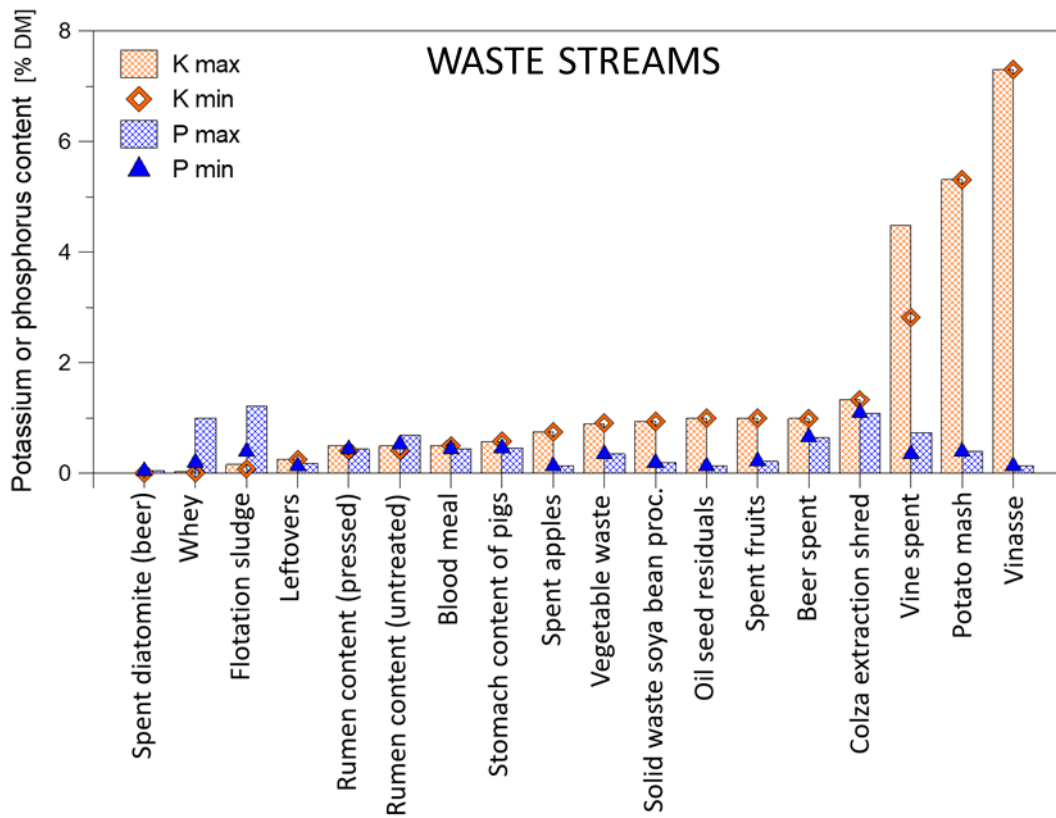


Fig. 22 Total phosphorus and potassium contents indicated as percentage of their DM content for different waste streams

D3.1. Classification of food waste and wastewater streams

Here, the waste streams with the highest phosphorus content are wheat mash, colza extraction shred from oil processing, beer spent and different streams from meat and animal feed production such as meat and bone meal, rumen content, stomach content and blood meal. Considering the actual phosphorus concentrations of those streams, the highest concentrations are also found in colza extraction shred from oil processing and in the meat and animal feed production waste streams (Fig. 20).

For the wastewater streams, the phosphorus concentrations range between <10 mg/L and 5200 mg/L (Fig. 21). Here, the highest concentrations were observed for wastewater from olive oil mills, soyabean processing, potato and potato starch processing as well as from dairies. However, as it is already known from soya bean processing wastewater for example, the phosphorus is unfortunately not always available as phosphate. In the untreated wastewater from soya bean processing only around 50% of the total phosphorus is available as phosphate (Moermann 2020). The main fraction of the total phosphorus is bound in phytic acid. In order to increase the phosphate fraction pretreatments such as an enzymatic phosphate release and/or anaerobic digestion are suitable. Moermann (2020) successfully increased the phosphate fraction due to an enzymatic phosphate release between 70% and 98% in his experiments.

In terms of potassium, the contents range from almost 0% to 7.5% DM. Here, the waste streams vinasse, potato mash, vine spent and colza extraction shred have the highest potassium contents related to their DM content (Fig. 22). Considering the concentration of potassium in the waste streams, the already mentioned waste streams are also relevant together with oil seeds residuals (Fig. 23). The potassium concentrations of those five streams are between 10 mg/L and 46 mg/L.

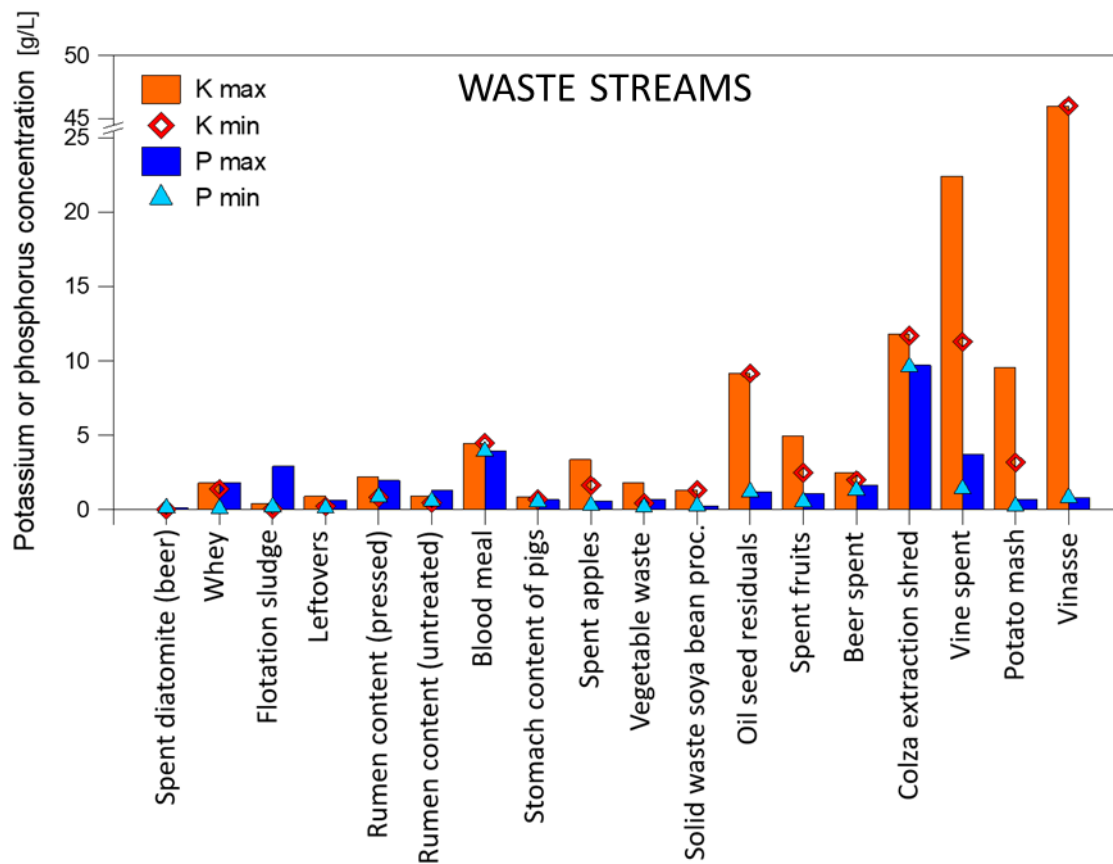


Fig. 23 Total phosphorus and potassium concentrations for the same waste streams as shown in Fig. 22 and in the same sequence.

For the recovery of K-struvite however, the phosphorus concentrations are important and should at least reach a certain level of 50 mg/L and higher (Cornel and Schaum, 2009). Even though, the phosphorus concentrations in Fig. 24 look very low compared the potassium and nitrogen concentrations, they range at a minimum between 240 mg/L for potato mash and almost 10 g/L for colza extraction shred. Thus, this might be still high enough for an economical rewarding P recovery. Furthermore, for the recovery of K-struvite, the concentration of ammonium needs to be so low (<1.1 mM corresponding to 20 mg/L) that the formation of NH₄-struvite will be avoided (Satoshi et al. 2013). However, Fig. 24 shows, that in all potassium rich waste streams at least total nitrogen concentrations between 3 g/L and 49 g/L are reported. Thus, if

potassium shall be recovered via K-struvite, either the nitrogen has to be depleted prior to the struvite formation or the formation of ammonium has to be avoided.

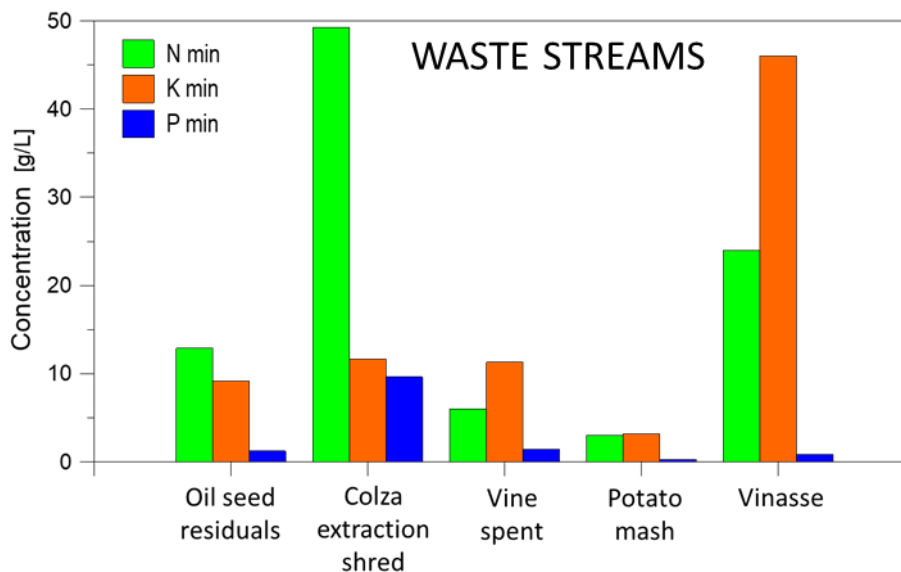


Fig. 24 Minimum concentrations from literature for total nitrogen, total phosphorus and potassium for potassium rich waste streams

Regarding potassium concentrations in the wastewater streams, there are only very few data available from literature. For wastewater from gelatine production, the concentration ranges between 20 and 190 mg/L (Silvano et al. 2018, Arturi et al. 2019), for wastewater from wheat starch production, it is around 450 mg/L and below (Bischofsberger et al. 2005) and for wastewater from olive oil mills, the concentration is between 3000 mg/L and 8000 mg/L (Waldron 2007). Own data from Moermann (2020) revealed a potassium concentration for wastewater from soya bean processing at around 1200 mg/L. The potassium contents of the waste stream shown in Fig. 22 and Fig. 23 suggest that after the degradation of the organic material potassium might be released and thus, being available in the liquor at least in a similar range as phosphate would be for most of the streams. However, this should be investigated in detail for the particular waste or wastewater streams. According to Bennett (2015), a minimum K:P ratio of 50:1 was found to be optimal for K-struvite precipitation. However, Moermann (2020) was already successful to produce K-struvite with a ratio of K:PO₄-P 15:1 corresponding to a K:P_{total} 6:1. In this case, only vinasse, potato mash and oilseed residuals with ratios of 44:1 and 10:1 and 6:1 would be in the range, respectively.

Conclusion for CIRCULAR ECONOMY regarding P rich (waste)water streams: struvite formation is possible & for K-struvite: N depletion prior to its formation is recommended

For phosphorus, quite high concentrations were found for wastewaters resulting from olive oil mills, soya bean processing, potato processing and dairies. Here, the concentrations are high enough for an economical awarding P recovery. The phosphorus contents in wheat mash, colza extraction shred, beer spent as well as waste from meat processing and animal feed production were the highest. However, usually, the phosphorus is not present as phosphate. Thus, a pretreatment is needed such as anaerobic digestion or an enzymatic treatment in order to release the phosphate.

The data suggest, that especially in vinasse, potato mash and oil seed residuals would have enough potassium for K-struvite formation. However, its nitrogen content is also quite high. If the formation of K-struvite is desired, ammonium should be removed first from the liquor. In Chapter 5.4, a suitable concept therefore is described.

Summary and conclusion in terms of CE technologies

The characterization of the waste and wastewater streams revealed for all wastes and wastewaters the suitability for anaerobic digestion, since their biodegradability is very high as also shown by their methane yields. Due to their high contents in oDM and in their COD concentrations, the five waste and wastewater streams with the highest potential are listed in Tab. 6 and Tab. 7, respectively. Also for their nitrogen, phosphorus and potassium contents and concentrations,

the most promising waste streams are summarized in Tab. 6 and for the most interesting wastewater streams in terms of carbon and nutrient recovery in Tab. 7.

Tab. 6 Overview on the five best results from the waste streams for each component and for its potential for recovery: same color means same origin of waste

Waste streams with a high potential for recovery of carbon and nutrients			
Carbon $y_{CH_4} > 0.45 \text{ m}^3/(\text{kg oDM})$	Nitrogen $N > 20 \text{ g/L}$	Phosphorus $P > 3 \text{ g/L}$	Potassium $K > 4 \text{ g/L};$ $K:P=6:1$
Flotation sludge	Blood meal	Colza extraction shred	Vinasse
Oil seed residues	Colza extraction shred	Meat and bone meal	Potato mash
Spent hops	Spent hops	Blood meal	Oil seed residuals
Meat and bone meal	Vinasse	Flotation sludge	
Blood meal	Potato mash	Vine spent	

Tab. 7 Overview on the five best results from the wastewater streams for each component and for its potential for recovery: same color means same origin of waste

Wastewater streams with a high potential for recovery of carbon and nutrients			
Carbon $CSB > 25 \text{ g/L}$ Biodegradability > 70%	Nitrogen $N > 1000 \text{ mg/L}$	Phosphorus $P > 200 \text{ mg/L}$	Potassium $K > 1000 \text{ mg/L};$ $K:P=6:1$
Fish processing	Pectin production	Olive oil mills	Soya bean processing
Olive oil mill	Soya bean processing	Soya bean processing	
Wheat starch production	Rendering (animal feed production)	Potato processing	
Distillery	Potato starch production	Potato starch processing	
Potato starch production	Wheat starch production	Dairies	

In general, the waste(water) streams of the meat processing industry (indicated in red), of the plant oil industry (in yellow), of sugar industry (vinasse, in blue), of the soya bean processing industry (in orange), of the potato processing industry (in green) and of breweries (light green) are the most interesting industries for the recovery of C/N/P/K. Based on this outcome, in chapter 5.5, the European country with the highest production rate in this industry will be determined and its regional distributions will be investigated in order to give the developers of the CE technologies in Circular Agronomics ideas, where their potential clients might be found.

Based on the chemical compositions of the waste(water) streams, suitable concepts for the application of the CE technologies will be provided in chapter 5.1. to 5.4.

5 Concepts for waste(water) streams with high potential for nutrient recovery and the technologies developed in Circular Agronomics to recover carbon and nutrients

According to the identified classification of the waste and wastewater streams in chapter 4, four concepts will be presented in this chapter. The characterization of the waste and wastewater streams showed, that prior to the recovery of nitrogen, phosphorus and potassium, an anaerobic treatment should be considered in order to increase the necessary availability of ammonium, phosphate and potassium. The types of recovery of the four concepts are summarized in Tab. 8.

Tab. 8 Overview about the considered concepts

Concepts for the recovery of carbon, nitrogen, phosphorus and/or potassium					
Concept name	Carbon		Nitrogen	Phosphorus	Potassium
	Biogas	Soil conditioner			
5.1 CN	✓	✓	$(\text{NH}_4)_2\text{SO}_4$		
5.2 CNP(Struvite)	✓	✓	$(\text{NH}_4)_2\text{CO}_3$	Struvite	
5.3 CNP(Brushite)	✓	✓	$(\text{NH}_4)_2\text{SO}_4$	Brushite	
5.4 CNPK (K-struvite)	✓	✓	$(\text{NH}_4)_2\text{SO}_4$	K-struvite	

For every concept, certain waste and wastewater streams have a high potential according to the outcome of chapter 4. Those streams are indicated for each concept. Furthermore, the regional distribution of those streams is presented in paragraph 5.5 for each stream.

5.1 Concept for C & N recovery and measures against ammonia inhibition during anaerobic digestion

In chapter 4.2, different waste and wastewater streams were identified, which are likely to contribute to an ammonia or ammonium inhibition during anaerobic digestion due to their high nitrogen content. Those substrates are blood meal, colza extraction shred, waste and wastewater from biofuel production, spent hops, vinasse and potato mash. In order to avoid this kind of inhibition, a concept was elaborated to continuously remove the ammonium from the digestate and to recover it as ammonium sulfate (Fig. 25). In addition in chapter 5.5, the regional availability and distribution of the relevant substrates for the shown concept are presented.

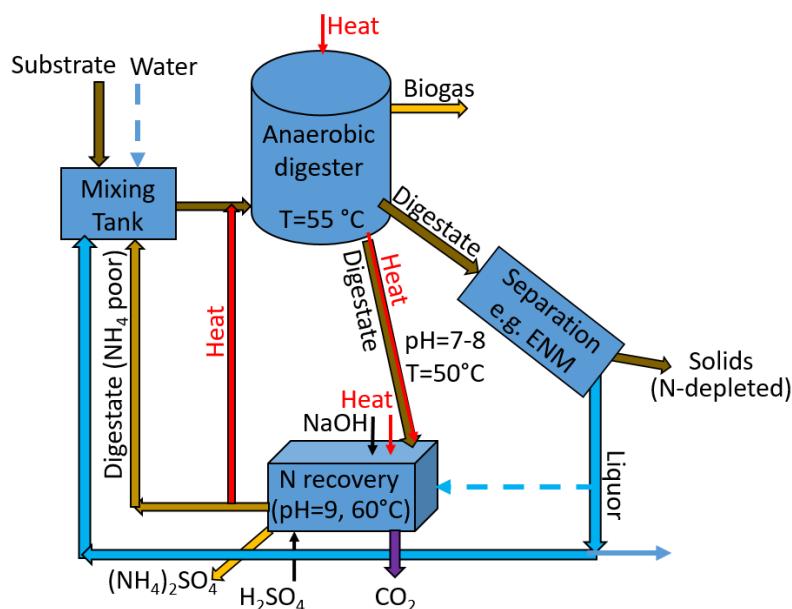


Fig. 25 Conceptual flowscheme for C & N recovery combined with the measure against ammonia inhibition

Most of the listed substrates are wastes and have high DM contents ranging between 20% and 98%. Hence, it is important to mix them either with water or with liquor prior to the digestion process in order to dilute their DM content. For the conventional anaerobic contact process (see also chapter 2), usually a lower DM content is required in order to provide good mixing conditions. In those digesters, the DM content usually ranges between 3% and 7%. The digestion process

can be either operated at conditions for thermophilic microorganisms at 55 °C or for mesophilic microorganisms at 37°C. A higher operation temperature however, might increase the sensitivity of the methanogens to an ammonia inhibition. Thus, the removal of the nitrogen is very important to avoid such an inhibition. For heating the digester content, due to economical and environmentally reasons, excess heat should be used, if it is available. For example, if the produced biogas is sent to a combined heat and power unit (CHP), the heat produced could be reused for that purpose.

For the removal and simultaneous recovery of the nitrogen, the organic bound nitrogen is converted into ammonium during the anaerobic digestion process. In a side stream, the recovery unit is implemented. There, the temperature is increased to 60°C and prior to the pH increase, CO₂ is degassed at vacuum conditions in order to decrease the necessary amount of NaOH for reaching a pH of 9.0. At those conditions, ammonium reacts to ammonia and can be degassed. It is injected in a scrubber operated with sulfuric acid in order to produce ammonium sulfate. This is a typical inorganic fertilizer ready to be used in agriculture. The N-depleted substrate is sent back to the mixing reactor for entering the anaerobic digester again. The heat contained in the N-depleted substrate might be also reused in order to heat the influent to the digester.

The resulting digestate is dewatered. Therefore, different technologies can be applied such as the ENM (see chapter 3), screw presses or centrifuges. The solids are N-depleted and can be reused in agriculture as soil conditioner. Furthermore, due to the digestion, easily degradable compounds are removed from the solids. Hence, they consist of stable organic carbon compounds. This is essential for a good soil structure and a good microbial activity in the soil.

The liquor resulting from the dewatering of the digestate can be sent back to the mixing reactor, or, if the ammonium concentration should still be in a high range, it can be sent to the N recovery unit. Concerning the liquor, other compounds and ions might accumulate, if it is circulated frequently in the loop. Therefore, sodium and potassium for example should also be monitored, their accumulation might also contribute to a decrease in the dewatering efficiency. If the accumulation of the ions approaches a critical level, a part of the liquor should be removed and/or exchanged with water. The dewatering efficiency might therefore serve as suitable parameter for monitoring.

5.2 Concept for C recovery, N recovery and P recovery inducing struvite formation

Depending on the type of the food and/or beverage industry, the composition of its wastewater will vary. Also, depending on the region and the origin of the used water for the production or processing of a certain product, the chemical composition of water varies in its calcium and magnesium concentrations. If the calcium concentrations are in a low range, e.g. below 100 mg/L (concluded from Chen et al. 2008), the following concept is suggested in order to recover carbon, nitrogen and phosphorus (Fig. 26). If the calcium concentrations are higher, the probability increases that precipitates occur at undesired points in the system. That might lead to clogging of pipes etc. Therefore, another concept is suggested in chapter 5.3.

The concept is for the anaerobic digester similar to that presented in paragraph 5.1. However, one difference is, that in the mixing tank enzymes can be dosed in addition. The enzymes are supposed to increase the release of phosphate from the organic material as described for the specific case of the treatment of soya bean wastewater in chapter 3.2. Furthermore, during anaerobic digestion, the phosphate and ammonium concentrations will further increase due to the anaerobic degradation processes of the organic material in the digester. After liquid-solid separation, the liquor is further treated for the removal of phosphate and its recovery as struvite. Therefore, in a first step, CO₂ is stripped in order to diminish the necessary NaOH demand for the increase in the pH to 8.0. With the addition of MgCl₂, struvite is precipitated and can be harvested. Struvite is a slow release fertilizer and can be applied in agriculture.

In a second step, the liquor is heated to 60 °C and the pH is further increased to 9.0 for the recovery of ammonia. The process is similar as described in paragraph 5.1. However, instead of using sulfuric acid for the production of ammonium sulfate, this concept suggests to reuse the CO₂ recovered via stripping in the P recovery unit. In the gas scrubber, as part of the N recovery unit, the CO₂ is dissolved in water and reacts partly to H₂CO₃. H₂CO₃ forms with the ammonia gas in the scrubber ammonium carbonate.

The resulting solids in this concept are expected to be not only N-depleted, but also P-depleted. Furthermore, the same criteria for its suitability as soil conditioner are here valid as already described in chapter 5.1. For the circulation of the liquor, also the same aspects as already discussed in chapter 5.1 should be taken into account.

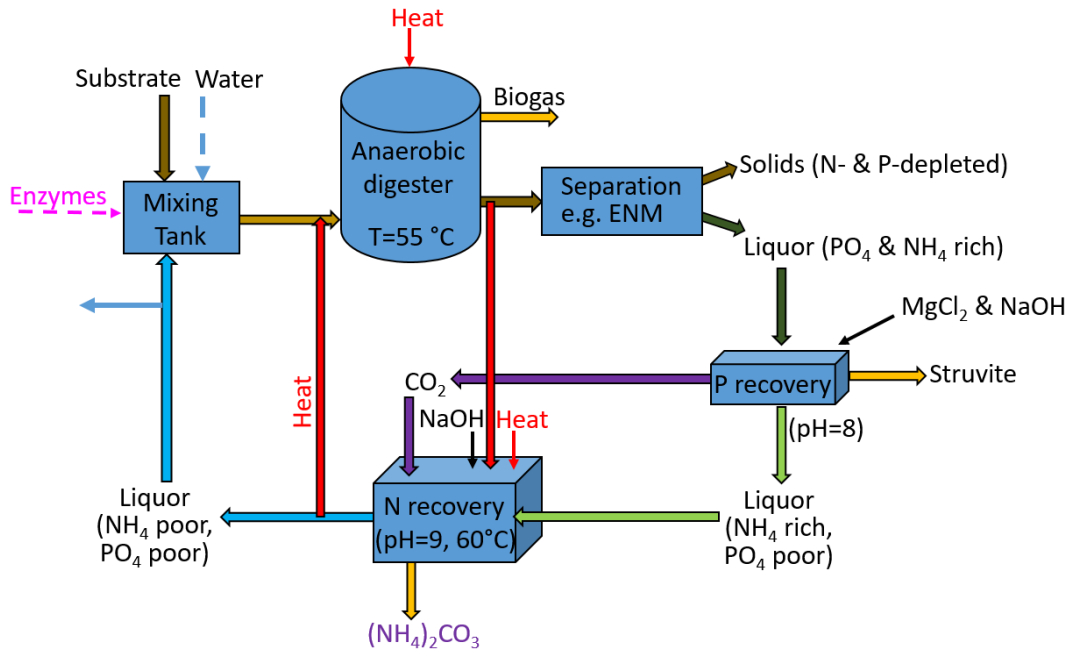


Fig. 26 Conceptual flowscheme for C, N and P recovery inducing struvite formation

According to chapter 4, suitable substrates for this concept might be colza extraction shred, oils seed residuals, waste and wastewater from biofuel production, waste(water) from meat processing and waste(water) from potato processing, because they have high carbon, nitrogen and phosphorus contents.

5.3 Concept for C recovery, N recovery and P recovery inducing brushite formation

In the case, the waste(waters) are rich in calcium (> 300 mg/L, concluded from Chen et al. 2008) or, if the tendency for undesired and uncontrolled struvite precipitations might be likely due to PO_4^- , Mg- and NH_4^- -rich waste(waters), a concept for P recovery via brushite formation is suggested as shown in Fig. 27.

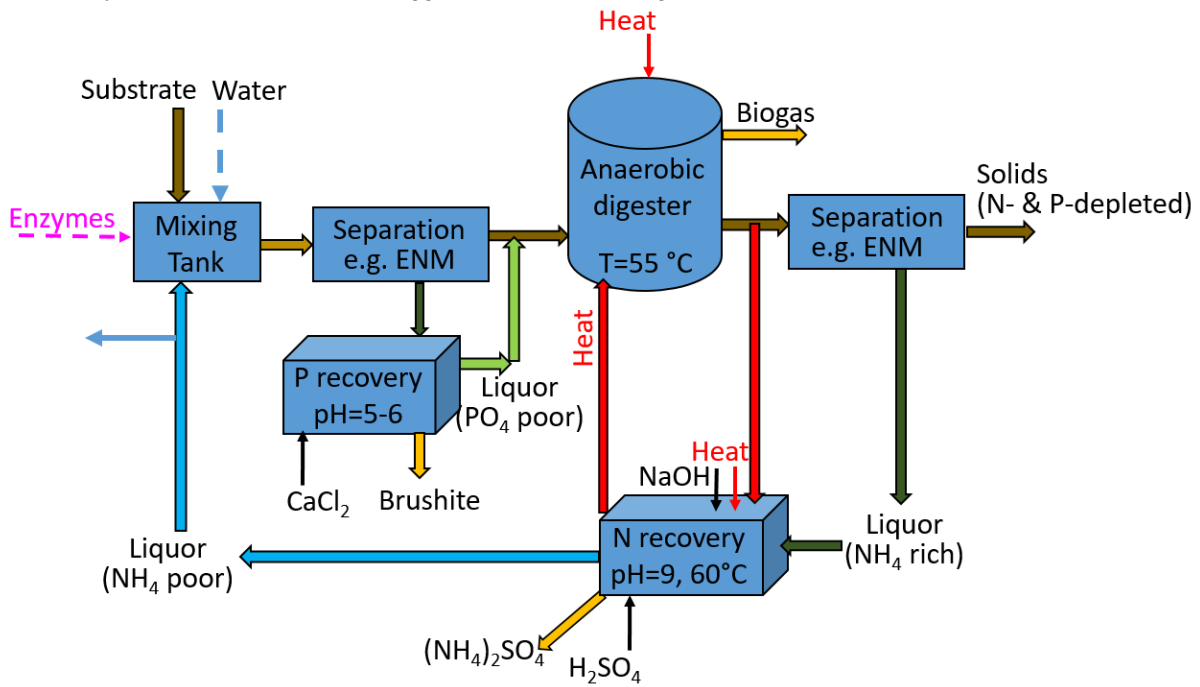


Fig. 27 Conceptual flowscheme for C, N and P recovery inducing brushite formation

Here, suitable substrates might be colza extraction shred, oils seed residuals, waste and wastewater from biofuel production, waste(water) from meat processing and waste(water) from potato processing (see chapter 4). As mentioned before, the fat containing substrates such colza extraction shred and oil seed residuals should be only digested with a carbohydrate rich co-substrate (see also chapter 5.1).

In the concept presented here, one difference compared to the concept inducing struvite formation in 5.2 is, that there are two dewatering steps necessary. The first dewatering steps takes place after the enzymatic treatment in the mixing tank, where phosphate concentrations are expected to be high. In the liquor, after the addition of CaCl_2 and if necessary, a pH adjustment to a level between pH 5 and 6, brushite is formed and can be harvested from the system. Brushite can be further used a fertilizer or as a food additive.

Regarding the N recovery system, the system works a described in paragraph 5.1. However, instead of treating streams containing solids, only the ammonium rich liquor is used here for ammonia removal via vacuum degasification. The vacuum degasification process is described in detail in chapter 3.2.

Hence, in this concept, in terms of C recovery biogas and an N- and P-depleted soil conditioner are produced. In terms of N recovery and P recovery, ammonium sulfate and brushite are produced.

5.4 Concept for C, N, P and K recovery

For substrates containing high potassium contents such as vinasse, potato mash and oils seed residuals, the recovery of potassium together with phosphate is considered (Fig. 28). However, because almost all waste and wastewater streams resulting from the food and beverage industry contain relatively high nitrogen contents as shown in chapter 4, it is necessary to remove ammonium prior to the K-struvite formation. Otherwise the K-struvite formation will not take place, because struvite will precipitate first as long as ammonium is available. Therefore, the concept suggests first to treat the liquor from the liquid-solid separation of the digestate via vacuum degasification as it is described in chapter 3.2. In order to diminish the NaOH demand for the pH increase to 9.0, the hot (60 °C) liquor is degassed for the removal of CO_2 . After the pH increase, also ammonia is degassed and reacts in a subsequent scrubber with sulfuric acid to ammonium sulfate. In the subsequent PK recovery unit, K-struvite is precipitated via the addition of MgCl_2 .

The heat management and the addition of enzymes as well as the avoidance of an undesired accumulation of e. g. sodium is recommended as already described in chapter 5.1 and 5.2.

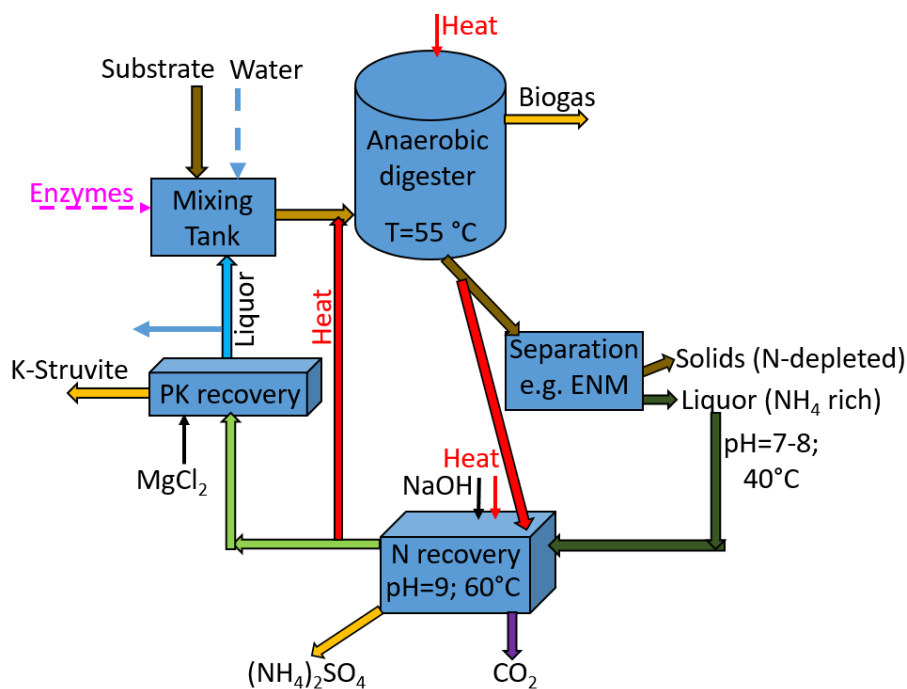


Fig. 28 Conceptual flowscheme for C, N and PK recovery inducing K-struvite formation

Hence, with this concept via C recovery, biogas and an N- and P-depleted soil conditioner are produced. In addition, via N recovery ammonium sulfate and via PK recovery K-struvite are produced.

5.5 Availability of waste(water) streams and their regional distribution

For the described concepts in the chapters 5.1 to 5.4, different substrates were indicated as especially promising for the recovery of the different nutrients as well as carbon in form of biogas for energy recovery and in the form of organically nutrient depleted material for soil conditioning. In the following subchapters the availability of those streams and their regional distribution in order to find clients for the new technologies are presented, mostly taking the case of Germany as example.

5.5.1 Waste and wastewater from slaughterhouses and meat industry

The waste from the meat industry such as flotation sludge, blood meal, meat and bone meal and animal feed production waste have very high carbon contents and high methane yields between 0.5 and 0.8 m³/(kg oDM). Furthermore, the nitrogen and phosphorus contents are quite high and thus, those streams are very well suited for the recovery of those components. The wastes and also the related wastewaters are found particularly in the meat processing industries and the slaughterhouses. Hence, the regional distribution of those sites was determined.

Considering Europe, the leading country in meat production is Germany, contributing 22% of the European meat production. Germany is followed by France, the United Kingdom, Spain and Italy with 19%, 14%, 11% and 9% (Eurostat 2009). Since Germany is the leading country in this industry type, the German market seems to be most promising for the promotion of the technologies developed in Circular Agronomics. Thus, the regional distribution of the industry for Germany was examined in Fig. 29. Four of the five biggest processing meat industries are located in the north west of Germany.

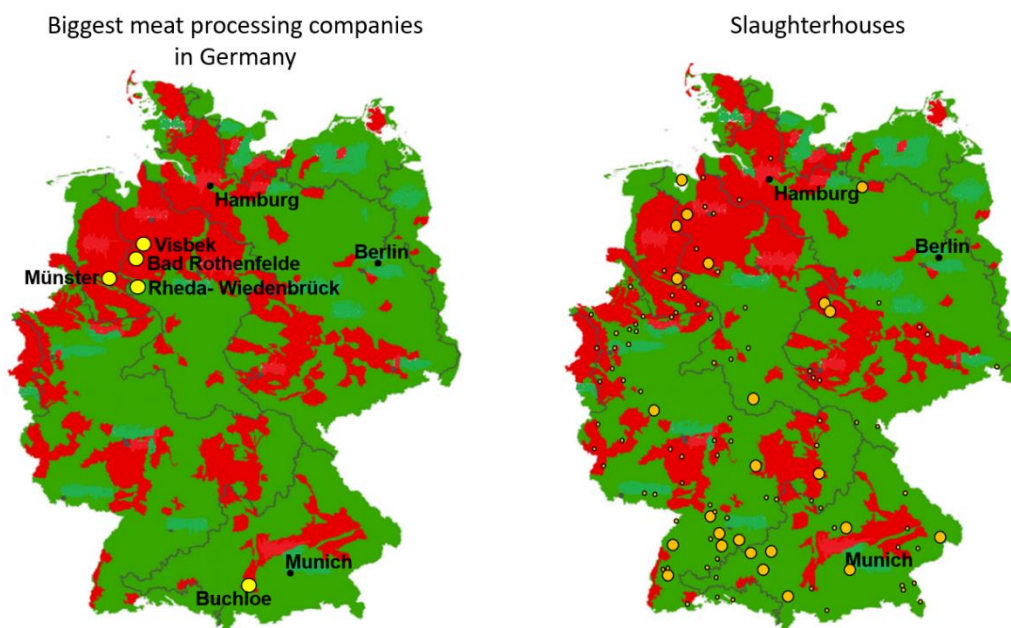


Fig. 29 On the left side, the biggest meat processing companies are shown according to data from Luo 2015 and on the right, the distribution of slaughterhouses in Germany is presented according to data collected by Kopf and Mayer 2020. The red marked areas are regions with elevated nitrate concentrations in groundwater, indicating excess nitrogen supply in agriculture (data from Bundesumweltamt 2017)

Here, the red marked regions suffer from high nitrate concentrations in the groundwater bodies indicating a nitrogen surplus in those regions. The application of organic fertilizers such as digestates and manure is highly restricted due to German Fertilizing Directive (DüV 2020). This directive was amended in 2017 and 2020 according to the European Nitrates Directive (1991/676/EEC) and the Directive on national emission ceilings for certain atmospheric pollutants (2001/81/EG). The directive limits the nitrogen and phosphorus amount for fertilizing in agriculture especially from organic fertilizers (DüV 2020). Consequently, the disposal of the nitrogen containing organic waste streams becomes more and more expensive in regions with a nitrogen surplus. Hence, there might be an interesting market for the proposed concepts, since they can contribute to diminish the disposal costs.

5.5.2 Waste and wastewater from colza oil industry

Colza extraction shred is a byproduct from the colza oil industry. It is produced during the extraction of the colza oil from the rapeseeds and it is gained via solvents such as hexane, which is later on removed from the colza extraction shred via a thermal treatment. Usually it is digested or used as animal fodder.

For digestion of colza extraction shred, high methane yields between 0.45 and 0.55 m³/(kg oDM) are reported. However, colza extraction shred and also oil seed residuals should be used as co-substrates and not be digested alone in a mono-fermentation process in order to obtain a stable biogas production process. Further substrates should be co-digested such as carbohydrate rich flow streams. Colza extraction shred and oil seed residuals have usually high contents in nitrogen, phosphorus and potassium as shown in chapter 4. Thus, those streams have a high potential for the application of the presented nutrient recovery concepts.

In Europe, the leading country in colza production and processing is Germany with a production of over 10,000 TJ/a followed by France, Poland, Czech Republic, Hungary and Bulgaria ranging between 1,000 and 10,000 TJ/a. (BLE 2017). Due to the high production rate in Germany, the regional distribution of the colza processing industries and the oil mills was determined according to BLE (2018a) in Fig. 30.

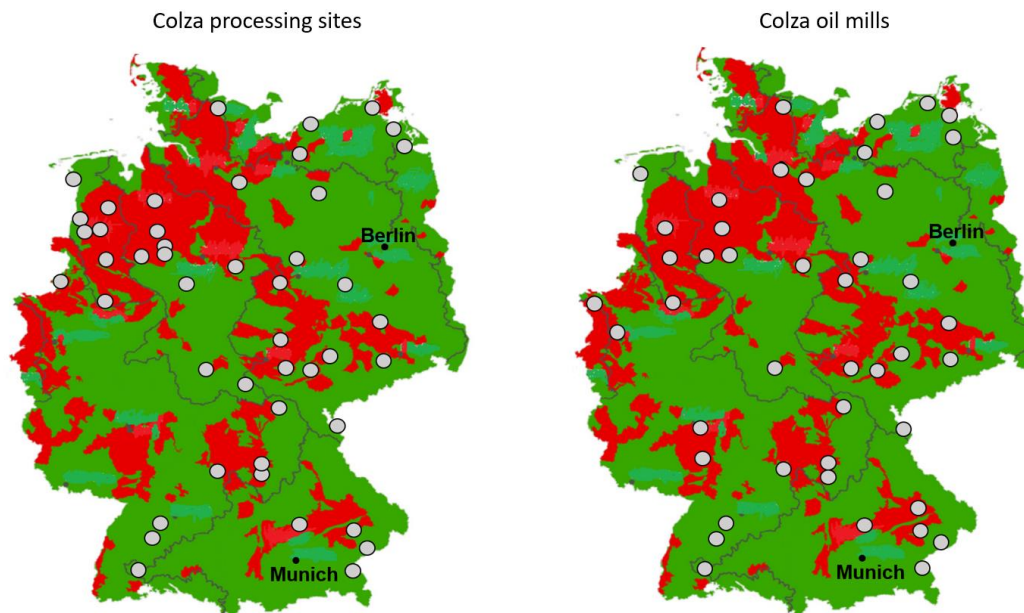


Fig. 30 Regional distribution of the colza processing industries (left side) and the colza oil mills (right side) in Germany (according to BLE 2018a) The red marked areas are regions with poor groundwater conditions (e. g. high nitrate concentrations; data from Bundesumweltamt 2017)

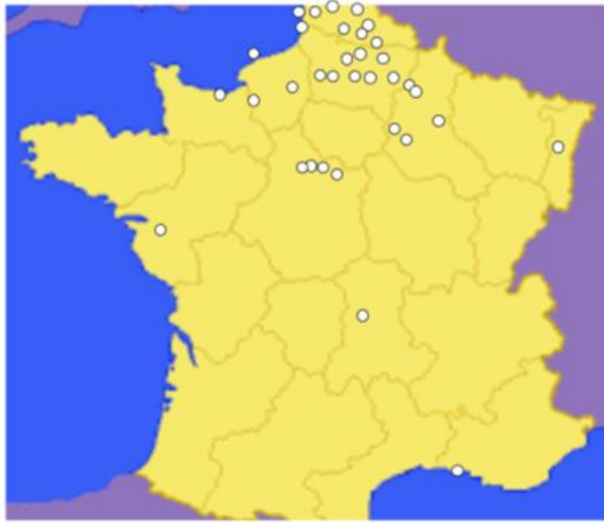
Corresponding to the regional distribution of the industries, the waste and wastewater resulting from the processes applied there have to be treated. Hence, at this sites, potential clients for the technologies developed in Circular Agronomics might be found. As already stated in paragraph 5.5.1, especially in the red marked areas, the increasing disposal costs of those waste streams might lead to the interest of those industries in nutrient recovery concepts.

5.5.3 Vinasse from sugar industry

Vinasse is a byproduct of the sugar industry. Usually it is digested or directly used as fertilizer on agricultural fields. However, due to the German Fertilizing Directive (DüV 2020), it is more complicated to apply organic wastes as organic fertilizer to the agricultural fields. The application is very limited by certain amounts and seasonal restrictions. Furthermore, usually, the time of application of organic fertilizer does not match the seasonal nitrogen demand of plants. Thus, the decoupling of the nitrogen supply of the plants and the carbon supply for the soil will contribute to turn the disposal into a valorization strategy. In Europe, France is the leading country for sugar production with 5 million t/a, followed by Germany, Poland, the United Kingdom and the Netherlands with 4, 2, 1 and 1 million t/a, respectively (Zuckerverbände 2020). Fig. 31 shows on the right side, the regional distribution of the sugar industry in France. Here, but also in Germany at the indicated sites, the byproduct vinasse is produced and if it is maybe already digested, the concepts presented here to

avoid ammonium and ammonia inhibition and furthermore the recovery of nitrogen might be very interesting for the operators.

Sugar production sites in France



Sugar production sites in Germany

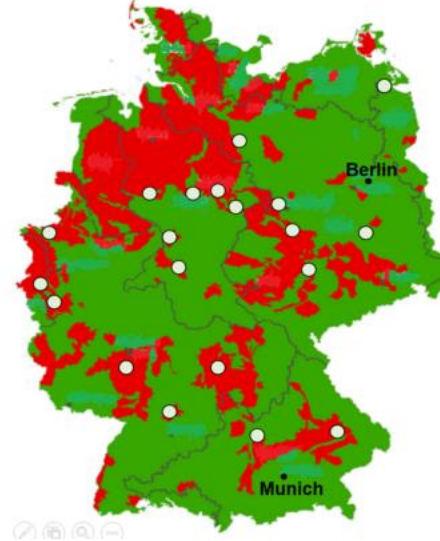


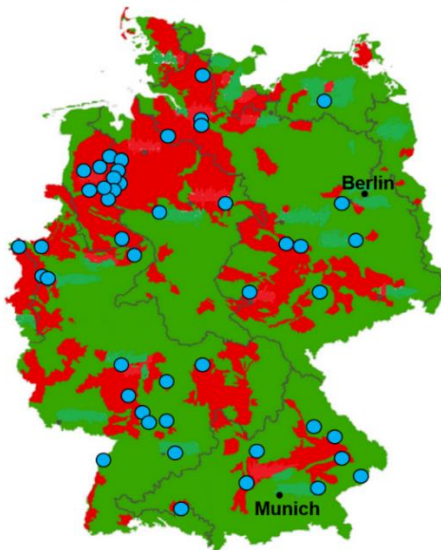
Fig. 31 Left: regional distribution of the sugar industry in France (according to data from IndustryAbout 2019); right: map of Germany showing the red areas which are regions with poor groundwater conditions (e. g. high nitrate concentrations; data from Bundesumweltamt 2017); indicating sugar producing industries. Most of them are located in or very close to the red areas.

5.5.4 Waste and wastewater from soya industry

As shown in chapter 3.3, wastewater from the soya industry is very well suited for the recovery of either struvite or even K-struvite. In Europe the leading country in soya bean processing is Spain followed by Italy, Germany, France and the Netherlands with 5,200 MT/a, 4,300 MT/a, 4200 MT/a, 3700 MT/a and 2500 MT/a, respectively (IDH 2017). Thus, it would be very interesting to show the distribution of the soya processing industries in Spain. However, unfortunately, according to the outcome of the internet research by the authors, those data seem to be not published.

Since the recycling of phosphorus is very much promoted in Germany and it is quite difficult in some areas in Germany to apply organic fertilizers to the agricultural fields, due to the German Fertilizing Directive (DüV 2020), its application and the availability of organic fertilizers is higher than their demand in some regions. Hence, the market in Germany for those CE technologies might be very promising. Therefore, the regional distribution in Germany is presented in Fig. 32.

Soya processing sites



Soya oil mills

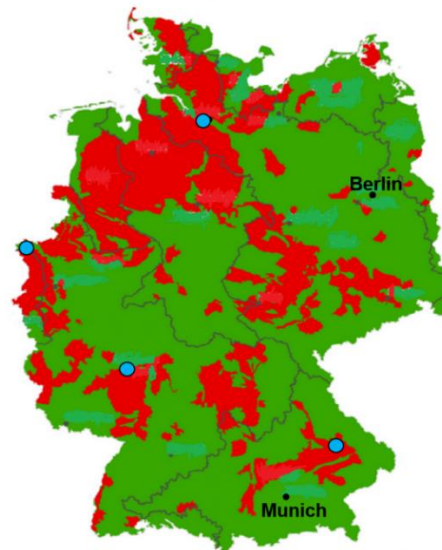


Fig. 32 Regional distribution of the soya processing industry in Germany (according to BLE 2018^a); the red marked areas are regions with poor groundwater conditions (e. g. high nitrate concentrations; data from Bundesumweltamt 2017)

At those sites, wastewater from soya bean processing is available and thus, there might clients for the CE technologies to be found.

5.5.4 Waste and wastewater from biofuel industry

The biofuel industry usually uses for the production of bioethanol cereals and sugar beets and for the production of biodiesel, colza, soya, palm oil and other plant oils (Braune et al. 2016). In chapter 4, the analysis of the different waste and wastewater streams showed, that the waste and wastewaters from plant oil processing and sugar production such as vinasse are very well suited for the recovery technologies proposed in the concepts in chapter 5.1 to 5.4. Due to the reason, that those material are also used in the biofuel industry, the potential for the recovery of carbon and nutrients seems to be high. Therefore, the regional distribution of those industries are shown here as well.

In Europe, the main producing countries of biofuels are Germany, France, the Netherlands, Spain and Poland with 140 PJ/a, 110 PJ/a, 80 PJ/a, 65 PJ/a and 40 PJ/a, respectively (Statista 2019). Because Germany produces the highest share in Europe, the regional distribution is the corresponding industries in Germany is shown in Fig. 33.

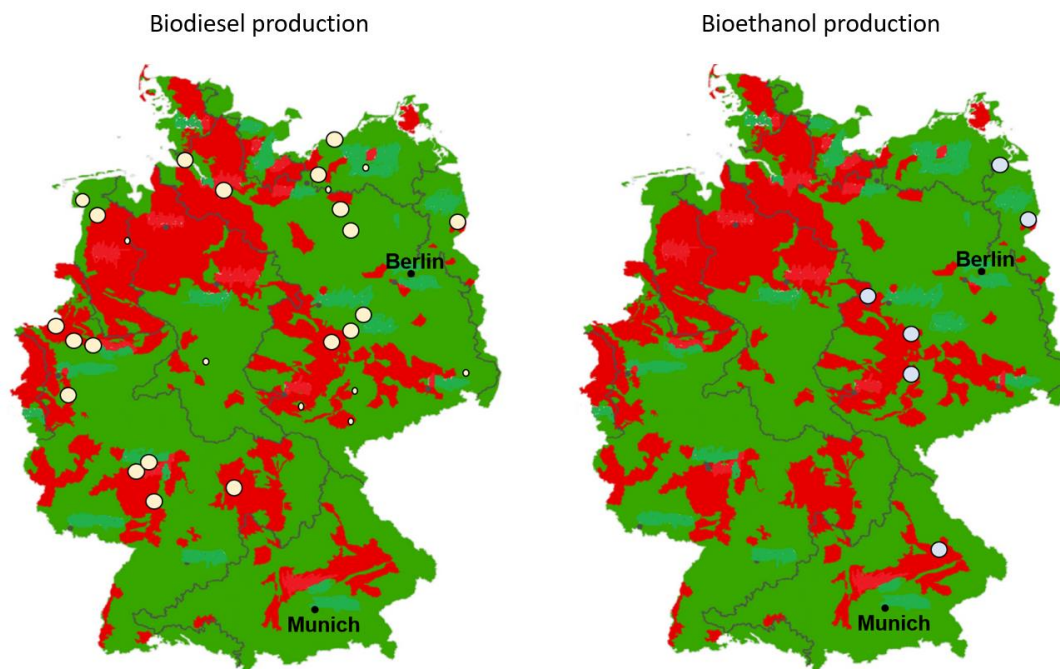


Fig. 33 Regional distribution of the biodiesel and bioethanol production industries in Germany (according to Braune et al. 2016); the red marked areas are regions with poor groundwater conditions (e. g. high nitrate concentrations; data from Bundesumweltamt 2017)

The biodiesel producing industry in Germany dominates with 28 sites compared to the bioethanol production with six sites. They are mainly found in the northern part of Germany. As already outlined in the previous paragraphs, the industries located in the red marked areas might be of special interest as a potential market for the proposed concepts.

5.5.5 Waste and wastewater from potato industry

Potato mash is a byproduct from the potato processing industry. Its methane yield ranges between 0.35 and 0.55 m³/(kg oDM) and thus, their digestion suggests to be economical rewarding. Also the wastewater from potato starch production has high COD, N and P concentrations and thus, is well suited for the recovery of the nutrients and the carbon in form of biogas as well as a soil conditioner for the digested solid material (see also chapter 4).

In Europe, the countries with the highest share of the produced potatoes are Germany, France, Poland, Netherlands and the United Kingdom with 17%, 15%, 14%, 12% and 10%, respectively (Eurostat 2018). Thus, one of market with the highest potential for the new CE technologies may be Germany. Therefore, the site for potato processing are shown in Fig. 34 according to BLE (2018b). For the potato starch producing industries, there are eight sites indicated mainly in the

northern part of Germany, while for the potato processing industries 14 sites are distributed all over Germany. Again, as already explained in the previous paragraphs, the industries situated in the red marked areas might be of special interest as potential clients for the proposed concepts.

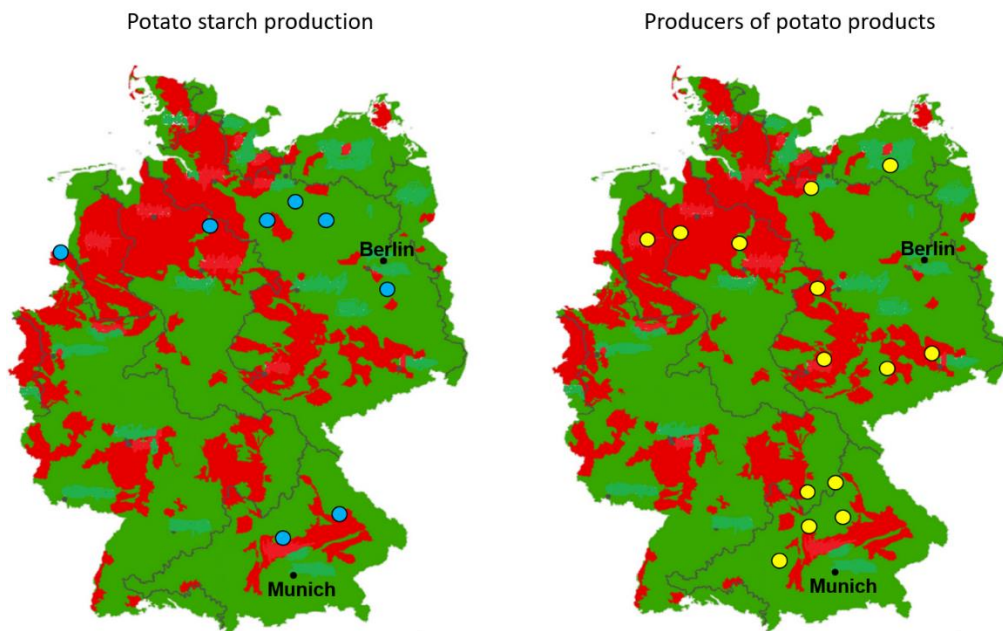


Fig. 34 Regional distribution of the potato processing industry in Germany: blue points mark starch production and yellow points mark producers of potato products (BLE 2018b). The red marked areas are regions with poor groundwater conditions (e. g. high nitrate concentrations; data from Bundesumweltamt 2017)

5.5.7 Waste and wastewater from breweries

Spent hops and beer spent are byproducts from the beer production process. Thus, the waste and wastewaters of breweries are promising flow streams for the suggested concepts for carbon, nitrogen and phosphorus recovery. According to chapter 4, the methane yields are between 0.35 and 0.55 m³/(kg oDM) for spent hops and beer spent. Also, the nitrogen and phosphorus contents are suitable for the application technologies.

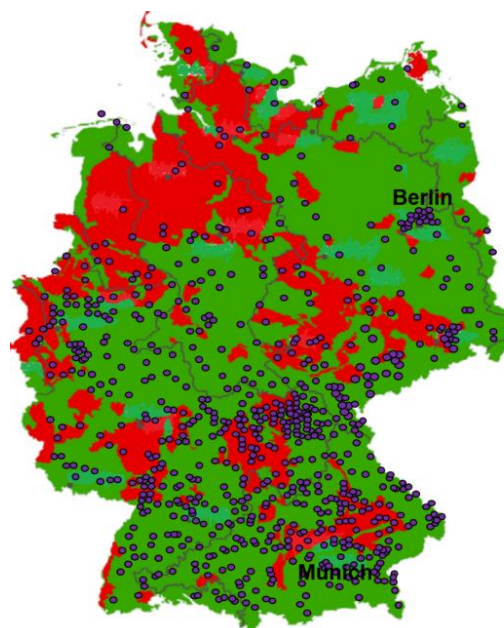


Fig. 35 Regional distribution of breweries in Germany (according to data published by Patzschke2020). The red marked areas are regions with poor groundwater conditions (e. g. high nitrate concentrations; data from Bundesumweltamt 2017)

In Europe the leading countries for brewing beer are Germany, the United Kingdom, Poland, Spain and Belgium with 8,300 million L/a, 4,500 million L/a, 4,000 million L/a, 3,600 million and 2,400 million L/a, respectively (Statista 2018). Therefore, the German market is very interesting for promoting the developed CE technologies, because it has the highest beer production in Europe. Fig. 35 shows the regional distribution of the breweries in Germany, indicating a high potential for clients mainly in the south of Germany.

6. Summary

In 2019, the European Commission published in its JRC Science for Policy Report the “Best available techniques (BAT) reference document for the food, drink and milk industries” also called BREF-document. In order to foster circular economy solutions in this sector, the current best available techniques in this document were reviewed and evaluated in terms of their suitability for closing the loops of water, nutrients and energy. This deliverable shows, that there is still a potential for new circular economy concepts to be integrated in this document. Even though this document lists already some treatment technologies which are very well suitable for circular economy solutions such as anaerobic digestion systems, nitrogen removal through ammonia stripping via aeration as well as phosphorus removal and recovery via struvite precipitation, some of the new technologies being investigated in the project Circular Agronomics are not listed yet in the document.

C recovery via anaerobic digestion for biogas production and stabilized organic material for soil conditioning

The characterization of different waste and wastewater streams from the food, drink and milk industry showed, that all considered streams in this deliverable are well suited for anaerobic digestion resulting in the conversion of organic matter into biogas. Furthermore, the anaerobic digestion process increases the phosphate and ammonium release due to microbial degradation processes. This is a beneficial pretreatment for the proposed technologies in terms of nitrogen and phosphorus recovery. In addition, the solid fraction of the digestate is already a stabilized material. Hence, easily degradable compounds are already removed from the solids. The solids are also mainly N- and P-depleted and can be reused in agriculture as soil conditioner. This is essential for a good soil structure and a good microbial activity in the soil and reduces severely the risk for emissions either as CO₂, N₂O or nitrate.

N recovery via stripping technologies

Even though the BREF-document includes only air stripping for ammonia removal as BAT, there exist more stripping technologies such as membrane stripping and vacuum degasification. The comparison of the three technologies showed for the vacuum degasification technology that no pretreatment is required, if the dry matter content does not exceed 7% in contrast to the other technologies. Due to that reason, the vacuum degasification has compared to the other technologies the lowest energy demand in this case.

Furthermore, the deliverable suggests the vacuum degasification system as a side stream treatment for anaerobic digestion of substrates with high nitrogen contents in order to prevent process failures due to a likely ammonium and/or ammonia inhibition of the methanogenic microorganisms. In contrast to the other stripping technologies, the milieu of the substrate remains under anaerobic conditions during the degasification process which supports the anaerobic requirements of the digestion process when the degassed side stream returns into the digester. Substrates with high nitrogen contents are especially blood meal, colza extraction shred, waste and wastewater from biofuel production, spent hops, vinasse and potato mash. Thus, the suggested concept might be interesting especially for the meat processing industry, the plant oil industry, biofuel industry, sugar industry, the soya bean processing industry and the potato processing industry and for breweries.

P recovery via struvite, K-struvite and brushite formation

Concerning phosphorus removal and recovery, also more concepts and technologies exist than listed in the BREF-document. For example, the combination with an enzymatic pretreatment in order to increase the phosphate release is up to now not suggested in the BREF-document. Especially for wastewater from soya bean processing, this deliverable describes three different treatment trains. The first two treatment trains recover struvite and the third treatment train recovers in addition to phosphorus also potassium as K-struvite:

- 1) phytase treatment of soya bean processing wastewater → anaerobic digestion → struvite precipitation → aerobic treatment;
- 2) phytase treatment of solid soya bean waste → either mixing with effluent from anaerobic stage of wastewater treatment or external ammonium addition → struvite precipitation → aerobic treatment
- 3) anaerobic treatment of the soya bean wastewater → aerobic treatment → K-struvite precipitation → tertiary P removal

Those treatment trains are also recommended to be included in the updated version of the BREF-document in the future.

D3.1. Classification of food waste and wastewater streams

The characterization of the waste streams suggested wheat mash, colza extraction shred, beer spent, meat processing waste and animal feed production waste for phosphorus recovery due to their high phosphorus contents. Additionally, the wastewater streams with the highest potential for phosphorus recovery result from olive oil mills, soya bean processing, potato processing, potato starch processing and dairies. For those (waste)water streams two concepts are described in the deliverable, producing either (1) struvite in combination with nitrogen recovery as ammonium carbonate or (2) brushite and ammonium sulfate. The applicability of the concepts depends on the chemical composition of the sludge and/ or liquor to be treated and its tendency to form uncontrolled precipitates such as struvite or calcium phosphate compounds at undesired positions in the treatment train. The suggested concepts might be of special interest for the meat processing industry, the plant oil industry, the sugar industry, the potato processing industry and for breweries. The (waste)water streams of those industries contain also high nitrogen contents as already described and thus, the phosphorus recovery is combined with the recovery of nitrogen in both concepts.

Due to the high potassium contents in vinasse, potato mash and oil seed residuals, the recovery of P and K as K-struvite is also suggested in a concept. However, prior to the recovery of the K-struvite, the ammonium has to be removed from the liquor via the recovery of ammonia resulting in the production of ammonium sulfate. This concept might be very interesting for the sugar, potato processing and plant oil industries.

*Application of **electrospun nanofibrous membranes (ENM)** in circular economy solutions*

According to Tili and Alkanhal (2019), the solid-liquor separation by using an ENM is a cost efficient alternative to a centrifugation step. Therefore, in all concepts, this technology is mentioned as an example for the dewatering step. However, up to now, that technology was mainly applied in laboratory testing stages and still lacks the experience in large-scale commercialization.

Therefore, this technology is investigated in detail in Circular Agronomics. Here, it is applied as a pretreatment of acid whey for thickening via nanofiltration. First experiments showed that the application of an ENM is a suitable alternative to centrifugation. However, up to now long-term experimental data are still needed to validate the hypothesis of an economical rewarding treatment in this case. This will be shown in the deliverable D3.2 available after February 2022.

Industries with a potential demand for the proposed concepts

In order to implement the proposed concepts, the research regarding the leading European countries in those industry sectors showed, that Germany has to highest production rates in the meat, colza oil, biofuel, potato and beer industries. In the sugar industry, France reaches the highest production rates followed by Germany. In the soya processing industry, Spain is the leading European country also followed by Germany. Furthermore, Germany is a very interesting market especially for the recovery of nutrients, because the direct application of organic fertilizers is restricted due to the German Fertilizing Directive (DüV 2020). Hence, especially in the regions with a nitrogen surplus, the disposal of the nitrogen containing organic waste streams becomes more and more expensive. This makes the industries in those areas to interesting potential clients for nutrient recovery concepts, since these can contribute to diminish the disposal costs.

Mostly for Germany as an example, since it has usually the highest production rates of all European countries, the regional distribution of the industrial sites is presented in the deliverable as a hint for the technology developer and providers, where their potential clients are located. For the meat industry, there are five main companies distributed over Germany. In contrast, the number of breweries in Germany exceeds 5000 and most of them are located in the southern part of the country. The potato processing industries are mainly located in the north of Germany. Colza, soya and sugar companies are almost evenly distributed over Germany. In France, the sugar industries are mainly found in the northern part of the country.

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Annex

Here, the technologies investigated in Circular Agronomics, are described in detail according to the required structure for the BREF-document. However, since the technologies are still under development, those descriptions are considered as a first draft. The authors suggest to update those descriptions at a later stage of the project prior to their potential integration in the BREF-document.

1. Centrifugation and nanofiltration for acid whey thickening in order to produce a substrate for biogas production

In dairies and milk processing industries, whey results as the remaining liquid after milk has been curdled and strained. Whey is distinguished in acid whey and sweet whey. Usually sweet whey is a by-product from the hard cheese production and has several commercial uses. In contrast, acid whey is often considered as waste and is often discharged into the sewerage system as for example in Slovenia, where 150,000 t/a are disposed to the sewerage system (Zupančič Justin 2020). In order to valorize the acid whey, it is thickened in order to be reused as biogas substrate and/or as soil conditioner. The valorization as biogas substrate is already practiced, however, the reuse as a soil conditioner is a new concept that will be investigated in detail in the project Circular Agronomics in work package 1.

1.1 Description of the treatment train or technology

The treatment train consists of two parts, the centrifugation unit and the nanofiltration unit (Fig. 36).

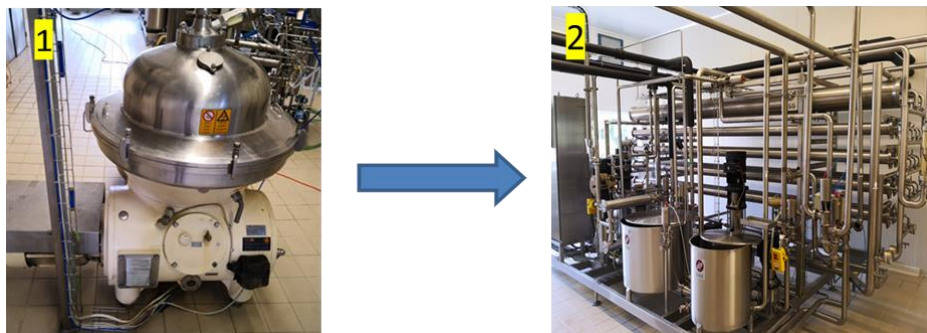


Fig. 36 Treatment train for acid whey thickening: (1) centrifuge and (2) nanofiltration unit

The nanofiltration unit (Fig. 36) is used for thickening of acid whey in order to increase the concentration of its macro components. As a filter, membranes from Dupont of the type DOW Filmtec NF 3840/30-F are used. They have a polypropylene outer shell and their pore size is approximately ~ 75 kDa. In order to protect the polymer membrane from fats contained in the feed whey, pre-centrifugation is used. The research results indicate that by centrifugation it is possible to reduce the fat content by 30% (Tab. 9). Thickening of the acid whey by applying the nanofiltration unit (NF) reaches normally a dry matter content of about 16 - 20%.

Tab. 9 Example: Parameters for the characterization of the feed (untreated) acid whey, the pretreated acid whey after centrifugation and the thickened acid whey after NF

	Feed acid whey	Acid whey after centrifugation	Acid whey after thickening
Fats, %	0.17 ± 0.02	0.12 ± 0.02	—
Non-dissolved matters, mg/l	485 ± 48	850 ± 85	1100 ± 111
Dry matter (DM), %	6.2 ± 0.6	4.7 ± 0.5	16.5 ± 1.5
Organic dry matter, % DM	91 ± 10	91 ± 10	91 ± 10
Inorganic dry matter, % DM	9 ± 1	9 ± 1	9 ± 1

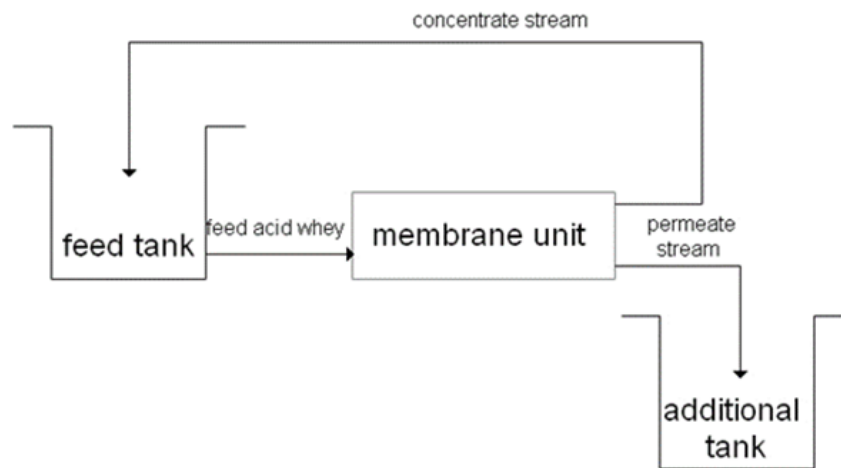


Fig. 37 Scheme of the pilot nanofiltration unit

Experiments for thickening acid whey were conducted with a feed flow of about 8,000 L/h and with a permeate flow of about 5,500 L/h. The retentate stream was recirculated to the feed vessel and the permeate stream was removed from the process (Fig. 37, Fig. 38). The experiments ended, when the feed solution had a dry matter content between 16% and 20%.



Fig. 38 Tank with the concentrated whey (a) and the purified stream (permeate)

1.2 Achieved environmental benefits

The acid whey feed is a waste from the dairy industry production. Usually, acid whey is diluted with water and disposed in the sewerage for further processing at the wastewater treatment plant. In order to foster circular economy, the described treatment train recovers valuable components from the acid whey. In addition, acid whey thickening allows producers to reduce waste production significantly. The concentration of acid whey by nanofiltration forms two streams: the treated one – permeate, and the concentrated one – retentate. The treated permeate stream can be reused to flush the nanofiltration unit and the retentate can be reused for other purposes, e.g. as substrate for biogas production and the subsequent application of the digestate as fertilizer and/or soil conditioner in agriculture. Furthermore, thickened acid whey (retentate from NF unit) can be applied as additive for animal fodder and as fertilizer for agricultural crops growth. The described treatment train thus recycles nutrients (N, P, K, C) from acid whey and brings them back into the nutrient cycle.

1.3 Environmental performance and operational data

This section will be updated later, when enough operational data are available. Therefore, the energy demands of the centrifuge and the nanofiltration (NF) unit will be considered. Furthermore, the water consumption for flushing of the centrifuge will be indicated as well as the permeate resulting from the nanofiltration which is used for flushing the NF unit.

For a reliable and long-term operation of the membrane elements, it is necessary to periodically carry out a cleaning in place (CIP) of the nanofiltration membranes. For chemical cleaning of the membranes, special solutions of acids, alkalis and surfactants are used.

1.4 Cross-media effects

The acid whey thickening technology allows to recover and reuse the dairy industry waste. Thickened acid whey can be used as substrate for biogas production, feed additive for animals in agriculture or for whey protein concentrate production.

1.5 Technical considerations relevant to applicability

The nanofiltration process is limited to a certain content of fats in the inlet stream of around 0.1%, since fat blocks the pores. Centrifugation is a well-established technology for the partial removal of fats. Requirements for the final product the thickened whey are determined by the dry matter content in a range between 18% and 22%.

1.6 Economics

For the calculation of economics, the OPEX of both stages for whey concentration (see 1.4.) will be considered. The OPEX result from the consumption of energy, water and chemicals. For the chemical cleaning of the nanofiltration unit, special solutions of acids, alkalis and surfactants are used. This section will be updated later, when enough operational data are available.

1.7 Driving force for implementation

Different companies in the dairy industry consider acid whey mainly as a waste product. In this case, the thickening of whey can reduce its amount by about three times, thereby reducing the amount of waste and the cost of delivering whey to waste treatment. With the additional use of whey, some economic benefits can be achieved: waste valorization instead of its discharge into wastewater treatment plants and paying fees; thickened acid whey can be valorized as substrate for biogas production, as a food additive for animals or by its application to an agricultural field.

1.8 Example plants

MADETA A. S. (Czech Republic)

2. Electrospun nanofibrous membrane and nanofiltration for acid whey thickening in order to produce a substrate for biogas production

An alternative for the treatment train for the valorization of acid whey as biogas substrate or as soil conditioner as outlined in chapter 3.1.1 is the replacement of the centrifuge in this treatment train by an electrospun nanofibrous membrane (ENM). According to Tili and Alkanhal (2019), the solid-liquor separation by using an ENM is a cost efficient alternative to a centrifugation step. However, up to now, that technology was mainly applied in laboratory testing stages and still lacks its large-scale commercialization.

In Circular Agronomics, this technology will be investigated in detail aiming to show, that it is economically and ecologically rewarding.

2.1 Description of the treatment train or technology

The treatment train consists of two plants, the ENM and pilot-scale nanofiltration units (Fig. 39, Fig. 40). The acid whey inlet stream is treated by nanofibrous membranes to reduce its fat content since its increased presence has negative effects on the membrane performance during nanofiltration (NF). The acid whey with a low fat content is pumped into the pilot-scale nanofiltration unit equipped with a membrane of the type DOW Filmtec NF 3840/30-F. The feed solution results then in the permeate stream and the concentrated stream called retentate. Experiments for thickening whey were conducted with a feed flow about 1000 L/h and at different membrane pressures. The retentate stream was recirculated to the feed vessel and the permeate stream was removed from the process (Fig. 41). The experiments were finished when the feed solution reached a dry matter content between 16% and 20%.

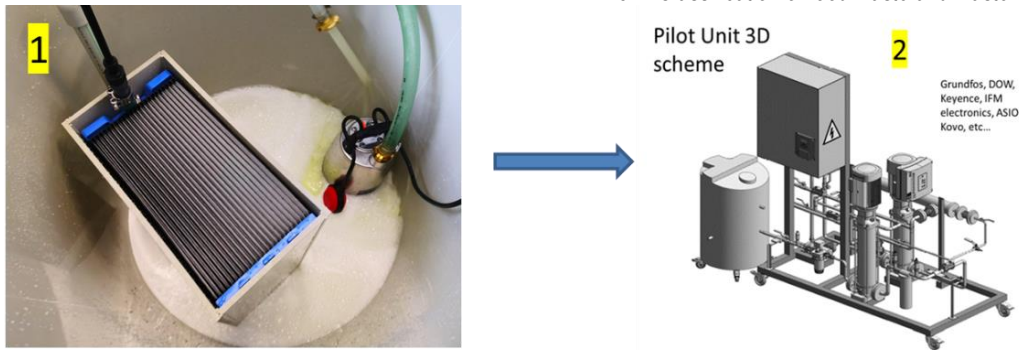


Fig. 39 Treatment train for acid whey thickening: (1) electrospun nanofibrous membranes and (2) nanofiltration unit

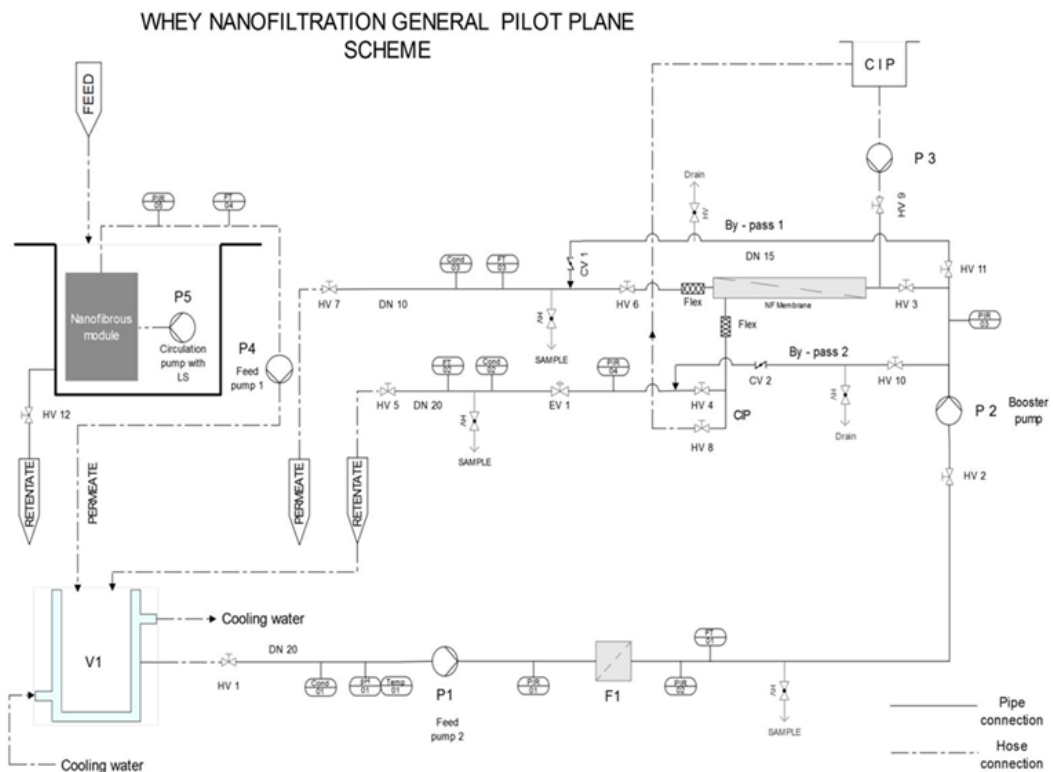


Fig. 40 Piping and instrument diagram (PID) of the pilot NF unit

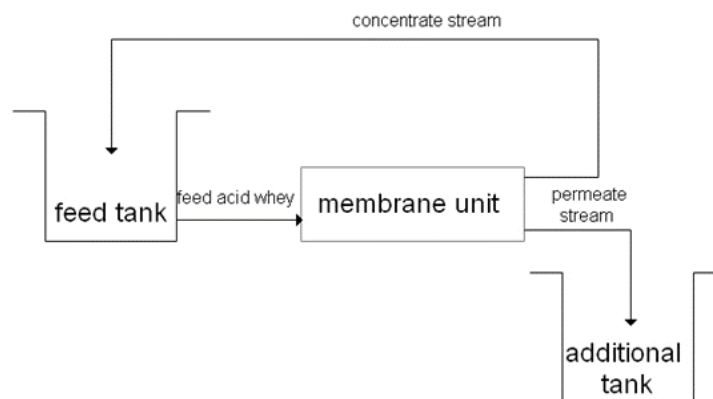


Fig. 41 Scheme of the treatment process for acid whey thickening via ENM and NF

2.2 Achieved environmental benefits

The acid whey feed is a waste from the dairy industry production. The acid whey thickening allows producers to reduce their waste production significantly. The concentration of acid whey by nanofiltration forms two streams: The treated

permeate stream can be reused to flush the nanofiltration unit and the retentate can be used in other areas, e.g. agriculture. Thickened acid whey (retentate from NF unit) can be applied for feeding animals and for agricultural crops growth. Acid whey is usually diluted with water and sent to the sewerage for further processing at the wastewater treatment plant. Our pathway thus recycles nutrients (N, P, K,) and carbon from acid whey back into the cycle.

2.3 Environmental performance and operational data (recovery rate, energy demand, chemical demand)

This section will be updated later subsequent document for the update of the BREF-document, when enough operational data are available. Therefore, the energy demands of the ENM and the nanofiltration (NF) unit will be considered. Furthermore, the water consumption for flushing of the ENM will be indicated as well as the permeate resulting from the nanofiltration which is used for flushing the NF unit.

2.4 Cross-media effects

Acid whey thickening technology allows to recover and reuse the dairy industry waste. Thickened acid whey could be used as food additive for animals in agriculture or for the whey protein concentrate production.

2.5 Technical considerations relevant to applicability (pretreatment, certain requirements)

The nanofiltration process can only be applied by a certain content of fats in the inlet stream, since fat blocks the pores. Central requirements for the fat content are a content of less than ~ 0.1% for the nanofiltration inlet stream. Those requirements arose from long-term performances of nanofiltration membranes.

The comparison of the pretreatment via membranes and centrifugation showed the same effectiveness of these stages in the removal of fats (Tab. 10). Thus, the centrifuge for the pretreatment of the NF inflow can be replaced by an ENM unit.

Tab. 10 Fat content of the feed (untreated) acid whey, the pretreated acid whey after centrifugation and the pretreated acid whey after ENM filtration

	Feed acid whey	Pre-treated whey after centrifugation	Permeate after ENM
Fats, %	0.17 ± 0.02	0.12 ± 0.02	0.1 ± 0.01

2.6 Economics

This section will be updated later in a subsequent document for the update of the BREF-document, when enough data are available. Therefore, the OPEX will be considered consisting of the consumptions of energy, water and chemicals. To compare the economics, the calculations were carried out separately for the stage of pretreatment and for the stage of thickening of whey.

2.7 Driving force for implementation

Different companies in the dairy industry consider acid whey mainly as a waste product. In this case, the thickening whey can reduce its amount by about three times, thereby reducing production of waste. Due to the reuse of whey, some economic benefits can be achieved. Instead of its discharge into wastewater treatment plants and paying fees, thickened acid whey can be valorized as a food additive for animals, as biogas substrate or applied to an agricultural field.

2.8 Example plants

There are no known full-scale application of acid whey thickening via ENM and NF at this stage. However, in Circular Agronomics, this treatment train is constructed as a pilot plant and operated, both by ASIO TECH, spol. s r.o. (Czech Republic). More information are available at the Circular Agronomics homepage or in D3.3 (to be updated after M42 (July 2022), when D3.3 is available).

3. Recovery of ammonia gas via vacuum degasification and gas scrubbing for ammonium sulfate production

In Circular Agronomics, different organic waste and wastewater streams from food and agricultural industry are investigated in terms of N recovery with vacuum degasification. Especially waste(water)streams with a high ammonium content such as digestates or manure are suited for this technology. The vacuum degasification unit is derived from a methane vacuum degassing unit for digested sewage sludge (TRL 9). In Circular Agronomics, this technology is further developed to an ammonia vacuum degassing unit (TRL 5) with a subsequent gas scrubber for the production of ammonium sulfate (TRL 9). Therefore, a pilot plant with a flow rate of 50 L/h is designed and constructed.

3.1 Description of the treatment train

The pilot plant consists mainly of two units, the vacuum degasifier and the gas scrubber (Fig. 42).

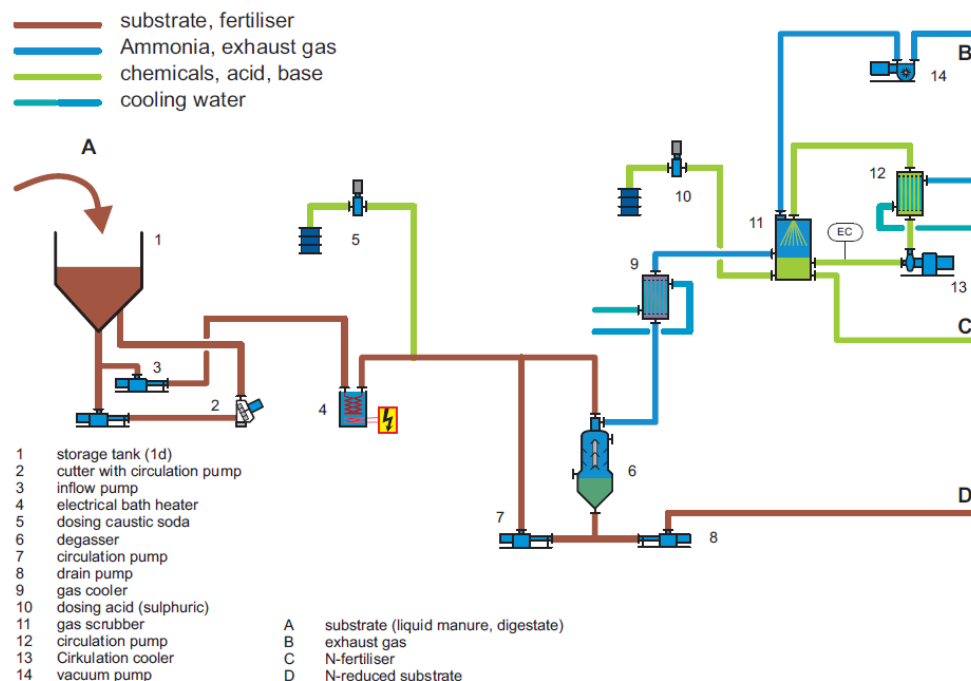


Fig. 42 Process scheme for vacuum degasification of ammonia gas with its subsequent scrubbing in sulfuric acid

The substrate is filled in the substrate tank, which is connected to a cutter in order to shred solids contained in the substrate. Substrates containing a total solid content of up to 7% can be processed in the pilot plant. The substrate stream can be circulated through the cutter as long as necessary until the solids are small enough for entering the heat exchanger and the degasifier. Here, if the pH is at neutral conditions, CO₂ can be removed via vacuum degasification. Furthermore, after the pH adjustment to alkaline conditions via NaOH and still at an elevated temperature, ammonium reacts to ammonia and can be degassed as well. The crucial process parameters such as pH, temperature and pressure can be varied in different ranges. In general, the pH will be adjusted between 8 and 10, the temperature can be varied between 20 °C and 70 °C and the pressure can be adjusted between 100 mbar and 900 mbar (indicated as absolute pressure). In the subsequent gas scrubber, sulfuric acid is circulated in order to react with the ammonia gas to ammonium sulfate. In agriculture, ammonium sulfate is also called ammonium sulfate solution. This is a typical mineral nitrogen fertilizer. After ammonia removal via degasification from the material, the so called “nitrogen depleted” material is ready to be applied as a soil conditioner.

3.2 Achieved environmental benefits

Digestates and manure are frequently used as organic fertilizers in agriculture, delivering organic material for the soil and nitrogen which is an important nutrient for plants. However, the seasonal application time of those digestates or manure is often not in line with the actual nitrogen demand of the plants. Consequently, an undesired loss of nitrogen for the plants due to emissions to the groundwater (nitrate) or to the atmosphere (ammonia and/or nitrous oxide) occurs and poses

serious environmental problems in regions with high rate digestate or manure application. The described treatment train enables the decoupling of the supply of organics and nitrogen contained in the digestate or manure.

3.3 Environmental performance and operational data

Circular Agronomics aims to achieve nitrogen recovery rates between 80% and 90% of the nitrogen which was originally present as ammonium in the digestate or manure. Thus, the application of nitrogen depleted material as carbon source for the soil as well as a demand driven fertilization of the fields will prevent the groundwater from nitrate emissions.

In order to reduce the demand for chemicals such as sodium hydroxide for increasing the pH in the fluid, CO₂ stripping of the fluid is recommended. Own experimental data showed, that for reaching and maintaining a pH of 9 at a temperature of 60 °C, the CO₂ stripping at that temperature and at a neutral pH saved 30% of the sodium hydroxide demand compared to the experiment without CO₂ stripping.

Concerning the energy demand, if the process is implemented at a biogas plant with a combined heat and power (CHP) system, the excess heat of the CHP should be used for the vacuum stripping process. This might even result in the reduction of more than 50% of the energy demand compared to that demand without the availability of excess heat.

3.4 Cross-media effects

The combination of that technology with anaerobic digestion is highly recommended due to the enhanced availability of ammonium/ammonia in the digestate serving as substrate for that technology. In addition, the reuse option of the excess heat from the combined heat and power (CHP) system saves energy and costs. More details are provided in the next two paragraphs.

3.5 Technical considerations relevant to applicability

Anaerobic digestion as a pretreatment in order to increase the degradation of organic bound nitrogen to ammonium is very favorable, since only nitrogen in the form of ammonium and further reacting to ammonia can be depleted via a stripping technology. Usually, the ratio of ammonium-nitrogen to total nitrogen in the raw wastewater is equal to 50% or less. For a higher and economically more favorable recovery rate in terms of nitrogen, the fraction of ammonium referring to the total nitrogen content needs to be increased. Therefore, anaerobic digestion of those wastewater streams can increase that fraction from 50% or less to a range between 60% and 80%. Further pre-treatment such as dewatering or filtering is not required.

3.6 Economics

The vacuum degasification reduces CAPEX due to its flexibility, since the total solid content to be degasified may comprise up to 7% and thus, there is no pretreatment such as dewatering or filtration necessary. If the process is implemented at a biogas plant with a combined heat and power (CHP) system, the OPEX might be reduced by more than 50% due to the usage of the excess heat of the CHP.

3.7 Driving force for implementation

Depending on the EU policy in terms of the actual execution of the Nitrates Directive within the European member states, the need for this technology might grow. Especially for areas with a high livestock density and hence, a high accumulation of manure as well as high concentrations of nitrate in the groundwater, this technology will help to deliver the necessary carbon compounds to the soil via the nitrogen depleted material, while simultaneously preventing the contamination of the groundwater via nitrate emissions. Furthermore, the nitrogen fertilizer produced by that technology can be applied, when it is needed and also where it is needed, since the costs for its transportation decrease due to the lower volume of the nitrogen fertilizer compared to the untreated manure or digestate. Also according to the new Fertilizing Products Regulation, from the summer in 2022 on, the nitrogen fertilizer produced by this technology will be officially labeled as a mineral nitrogen fertilizer.

3.8 Example plants

- Vacuum degassing unit for ammonia (TRL 5): Circular Agronomics homepage or D3.4 (in February 2022, when D3.4 is available)
- Vacuum degassing unit for methane (TRL9): <http://www.pondus-verfahren.de/>

4. Enzymatic enhanced phosphate release with subsequent struvite production

It is well known that some major food and feed substrates do contain phosphorus as phytic acid, a multiple P containing cycle molecular structure. One of the most important feed/food additives in this regard is soya bean. Phytic acid has a known limited degradation during anaerobic wastewater treatment. This renders phosphate (PO_4) recovery by means of struvite formation difficult. In the subsequent aerobic stage the degradation of phytic acid occurs, releasing the P as PO_4 and requiring significant dosing of ferric or aluminum salts to attain the final P discharge levels for the effluent. If phytic acid conversion into PO_4 could be enhanced during anaerobic processing, this would result in a wastewater composition suitable for struvite formation. In addition, analysis of compounds show that in this particular case PO_4 would be the limiting parameter rather than magnesium. The selected approach to reach higher PO_4 levels is to increase the phytase enzymatic activity by either added commercial available phytase enzymes or by cultivating in vivo high phytase producing fungi or yeasts.

4.1 Description of the treatment train

The pilot unit has a multiple purpose and can be run in different scenarios at an average flow of $1 \text{ m}^3/\text{h}$. Fig. 43 gives an overview of the general wastewater and solid waste process flow and how they are processed at this stage in the full scale soya bean processing plant. The possible interaction with added phytase enzymes to improve the conversion of bound-P into to recoverable PO_4 are indicated as option 1 and option 2.

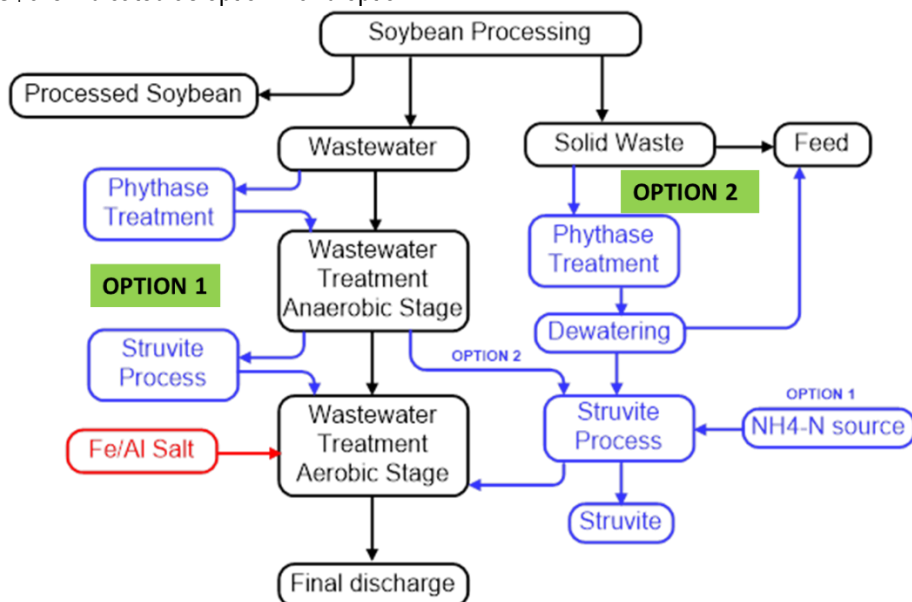


Fig. 43 Main stream processing of wastewater and solids wastes.

The major gain can be achieved on the main wastewater flow. The normal processing is a straight forward anaerobic treatment after buffering and equalization. The wastewater is at $40\text{-}50 \text{ }^\circ\text{C}$ and pH of $4.5\text{-}5.5$. This requires some cooling by active heat exchange. Given the low pH and elevated temperatures these conditions do favor selected growth of fungi/yeasts and are in line with optimal process conditions for a number of commercial available phytase enzymes. One should take note that this low pH and lack of free ammonium does prevent the struvite formation at this stage. The latter is also the case for possible K-struvite formation, the pH at this stage is too low.

In the adapted flow the wastewater is pre-treated with phytase enzymes. These enzymes are a bulk commodity already largely used as feed additive to improve digestibility. By adding the selected phytase enzyme(s) the phytic acid is hydrolyzed prior to entering the anaerobic stage. Since there is no or only little PO_4 uptake of the liberated phosphate, that will be incorporated into new anaerobic cell biomass (there is only limited sludge growth under anaerobic conditions), most of the produced PO_4 will remain in solution and be present in the anaerobic stage effluent. In addition, the anaerobic treatment will also convert organic compounds liberating the contained nitrogen as ammonium (major constituent of struvite) as well as increase the pH. Under these conditions, high levels of PO_4 , NH_4 and an elevated pH are reached and the formation of struvite can be induced. This results in phosphorus removal/recovery between the anaerobic and aerobic stage of the wastewater treatment. An alternative approach would be instead of adding commercial phytase enzymes, trying to induce in vivo phytase production in the buffer prior to the anaerobic stage. Since there are no indications that in

vivo phytase production occurs at this stage a separate cultivation reactor, feeding continuously phytase active fungi/yeasts in the buffer, might be needed to work according to this approach.

Finally a last possibility (option 2) is to investigate, if phytase treatment can release additional PO_4 contained in the separated solids.

Fig. 44 shows, how the different options 1 and 2 can be applied, and their locations within the existing treatment processes. The enzyme aqueous solution can be prepared separately starting from purchased enzymes as a powder and dosed at a given ratio versus the incoming flow into a separate hydrolysis reactor. The in vivo phytase activity can be obtained by cultivating selected fungi/yeasts strains in a separate correctly conditioned reactor (pH: 3.5 – 4.5 and temperature: 40-45 °C). The bypass of incoming wastewater can be used as feed source to induce phytase production. The in vivo reactor content can either be introduced as such in the phytic acid hydrolysis reactor or after the separation of the active biomass, the processed liquor can be used for that. These additional treatments on the wastewater flow are aimed at increasing the available PO_4 which at an elevated pH and with liberated ammonia can be converted into NH_4 -struvite. The latter can be done by either a pH increase (given the high level of magnesium (Mg) already present in the wastewater) or by adding extra Mg to achieve higher recovery rates. Processing the solids is the second option. A blended mixture of these solids and phytase enzymes (commercial product) can be incubated to release additional PO_4 . An extra dewatering phase would thus generate extra PO_4 for struvite formation.

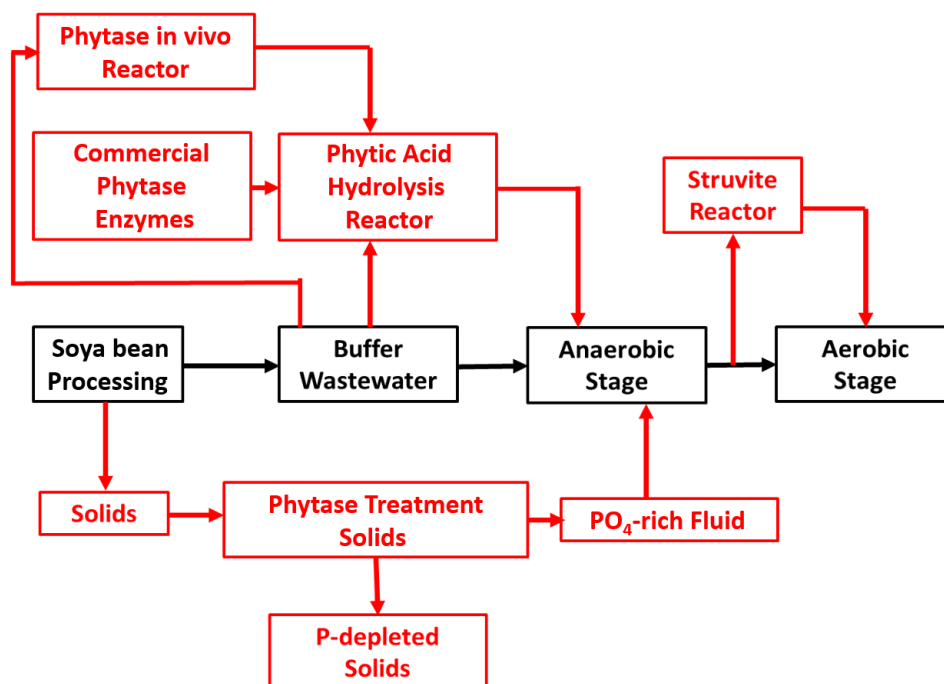


Fig. 44 Black flow is the current treatment/ red flow is the proposed possible phytase treatments

4.2 Achieved environmental benefits

The following advantages are linked with struvite formation in wastewater and are related to the management of the phosphorus mass balance. Phosphorus needs to be extracted as a solid either immobilized in the aerobic biomass or precipitated with mainly iron (Fe-) or aluminum (Al-) salts. Those salts generate an extra quantity of chemical sludge to be processed further. In addition, the phosphate removed in Fe/Al-precipitates is hardly any longer available as plant nutrient. Another not negligible side effect is an increase in salinity due to the counter-ions of Fe- or Al-salts used.

The alternative of struvite production to precipitate phosphate (more in particular crystallize) results in compounds containing two major plant nutrients, nitrogen (N) and P, with some added Mg. Mg is an important micro-nutrient as it is the key element of chlorophyll. An important feature of struvite is its low water solubility resulting in slow release properties as a fertilizer. The release of the contained N and P is higher in acidic soils, but another important process is the active

nutrient extraction of plants by their root system. Hence, besides having a process capable of converting the soluble PO_4 into a solid state product, a reusable fertilizer (or fertilizer additive) is produced.

Very specifically for this type of wastewater is that Mg is not the limiting parameter, but PO_4 is. This opens a unique opportunity to produce struvite without any addition of Mg due to a simple pH control which can be induced by CO_2 stripping combined with a strict pH control of the anaerobic stage. Thus, the benefit is a phosphate removal combined with nutrient recovery that only has some extra energy demand (aeration).

4.3 Environmental performance and operational data

The struvite recovery rate depends on the process conditions and is determined by the concentrations of the parameters involved which determine the saturation ratio. The pH of the wastewater is a major process parameter. When conditions are correct the final chemical equilibrium results in residual PO_4 levels of 20-30 ppm $\text{PO}_4\text{-P}$. The initial P concentration in the incoming wastewater is 120-140 mg P/L. So, the recovery rate will be in the range of 75-85 %. The energy demand will depend on mixing energy or aeration turbulence in this particular case. At an air/wastewater ratio of 5-7 the air requirement is 625 – 875 Nm^3 (10-15 Nm^3/m^3 wastewater), this can be generated by an 8-12 kW blower. As Mg is already available, there is no need for extra Mg. The only product that has to be added (if not cultivated in vivo) is the commercial phytase enzyme at dosing rates of 5-10 mg/kg COD or 3 to 6 kg/day.

4.4 Cross-media effects

This technology approach has a beneficial effect on the residual salinity and reduces the use of chemicals to a minimum. This is due to the fact that PO_4 is the limiting parameter for this particular type of wastewater. Furthermore, phytase enzymes are readily available as chemical commodity product and come in a wide variety of enzymes each with specific optimum pH and temperature. So any type of wastewater rich in phytic acid could be subjected to a similar treatment.

4.5 Technical considerations relevant to applicability

The phytase treatment is done prior to the anaerobic treatment of the wastewater. The option of dosing the enzyme are discussed in 1.2. The hydrolysis of the phytic acid can be done in a separate reactor. The treatment requires a 3-4 hour period to get an enough high conversion rate. However, this rate can probably be increased by dosing more enzymes. Due to the low consumption, this is a viable option. In most cases the existing buffer tank is positioned prior to the anaerobic stage that can be used to dose the phytase enzyme. If not already present, a mixer is required to get a good uniform distribution of the added enzyme.

The second major technical adaptation is the integration of a struvite reactor. In this particular case a combined air stripping/crystallization will be needed. This would mean a reactor size of 150 m^3 combined with an aeration system and a pH control.

4.6 Economics

The OPEX are related to enzyme treatment. The highest demand in the studied case was 10 kg enzyme/day. This would account to less than 100 €/day for enzyme costs. The amount of FeCl_3 which is correspondingly saved is estimated with the $\text{Fe}/\text{PO}_4\text{-P}$ molar ratio of 1.5 (on 70% of P load) and is about 2000 kg of FeCl_3 resulting in 1000 €/day. Also, this amount of FeCl_3 added will result at least in a 1700 kg DM/day. Processing and disposal of the extra sludge are 75 €/ton or at 20% DM this accounts up to 640 €/day. So chemical savings and the related saved costs are about 1640 €/day (0.5 to 0.6 million €/year).

The CAPEX will contain the implementation of a mixer (if not present yet with around 30,000 €) and the struvite reactor. This struvite reactor contributes as a major factor to the costs and is estimated (for the size of 125 m^3/h) at 0.75 to 1 million €.

4.7 Driving force for implementation

Besides the possibility of recovering phosphate and the production of a reusable fertilizer or fertilizer commodity product as well as the avoidance of disposing of phosphorus in a non-sustainable way, there are also other incentives to use this approach: reducing the salinity in the final effluent which renders its reuse as irrigation water more likely. This type of water supply will become more and more prominent in the coming years due to climatic changes. However, the main driver is in

this particular case, that very low dosing rates of enzymes and the fact that sufficient Mg is present (due to vegetable origin of the substrate), the bound phosphorus in the phytic acid can be converted into PO_4 which is mandatory, if struvite formation is wanted.

4.8 Example plant

There are no known full-scale application of phytase enhanced struvite recovery at this stage. However, in Circular Agronomics, this kind of struvite production plant is constructed and operated. More information are available at the Circular Agronomics homepage or in D3.3 (to be updated after M42 (July 2022), when D3.3 is available).

5. Enzymatic enhanced phosphate release with subsequent K-struvite production

Phosphorus related issues have been described already in 3.3.1. *Enzymatic enhanced phosphate release with subsequent struvite production*. Since phosphorus is a limited resource and essential for food/feed production, a fertilizer reuse will be mandatory to ensure food safety in general for future generations. Besides nitrogen, the other key fertilizer compound is potassium. Also, potassium is a mined resource with limited supply. Potassium is as opposed to N and P not a major constituent of biomass, but it is a key element in terms of metabolic activity and ensures good cation/anion equilibria and transmembrane transport mechanisms. The latter is inherently coupled with the high water solubility of K. This high water solubility is an extra challenge, when focusing on K-recovery since the first step in any type of recovery processes is most likely an extraction process in order to obtain a concentrated flow out of a diluted aqueous flow. A compound with low water solubility renders the latter feasible. One of the few K-salts with low water solubility is K-struvite (the analogue to main stream NH_4 -struvite). The general rule is that as long as NH_4 is present NH_4 -struvite will be produced, only when NH_4 is depleted and if at that moment still PO_4 is available, K-struvite will be formed. In addition, K-struvite requires a higher pH compared to NH_4 -struvite under similar conditions.

5.1 Description of the treatment train or technology

For the production of K-struvite, NH_4 needs to be depleted. Thus, K-struvite formation is only possible after the aerobic stage. The wastewater has a potassium concentration between 1000 and 1500 ppm that is also observed in the aerobic stage effluent. A part of the dissolved Mg in the effluent from the anaerobic stage precipitates in the aerobic stage, but the other part will still be dissolved and available in the aerobic stage effluent. The key parameters to control the precipitation process are the pH and, if needed, additional dosages of Mg. Fig. 45 shows the implementation in an existing plant. The technology train will require a struvite crystallization reactor but also a change in the overall P management. If the goal is K-struvite formation after the aerobic stage, no addition of Fe/Al in the aerobic stage is allowed in order to maximize the PO_4 throughput towards the K-struvite unit. Also the final PO_4 obtained after K-struvite formation will be in order of 20-30 ppm PO_4 -P. Hence, there will be the need for an additional tertiary P-removal after the K-struvite production in order to attain the required final discharge levels. In most cases, a type of tertiary P-removal is already implemented. The K-struvite process is applied on the entire flow of the wastewater.

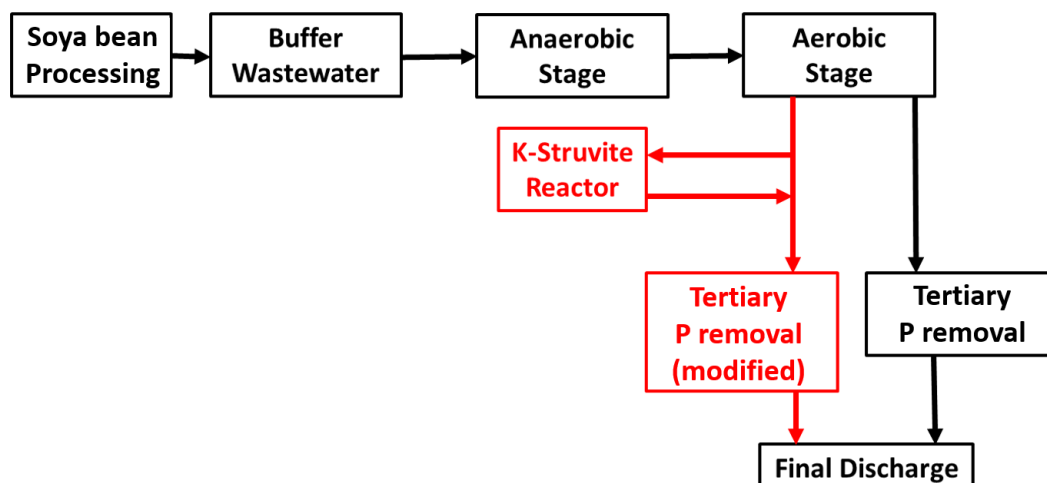


Fig. 45 Implementation of the K-struvite process in an existing plant

5.2 Achieved environmental benefits

The following advantages are linked with K-struvite formation in wastewater and are related to the management of the phosphorus mass balance. Phosphorus needs to be extracted as a solid either immobilized in the aerobic biomass or precipitated with mainly Fe- or Al-salts. Those salts generate an extra quantity of chemical sludge to be processed further. In addition, the phosphate removed in Fe/Al-precipitates is hardly any longer available as plant nutrient. Another not negligible side effect is an increase in salinity due to the counter-ions of Fe or Al-salts used.

The alternative of K-struvite production to precipitate the phosphate (more in particular crystallize) results in compounds containing two major plant nutrients, K and P, with some added Mg. Mg is an important micro-nutrient as it is a key element of chlorophyll. An important feature of K-struvite is its low water solubility resulting in slow release properties as a fertilizer. The release of the contained K and P is higher in acidic soils, but another important process is the active nutrient extraction of plants by their root system. Hence, besides having a process capable of converting the soluble PO_4 into a solid state product, a reusable fertilizer (or fertilizer additive) is produced.

The reported environmental benefits are identical with the benefits related to § 4. *P recovery via enzymatic enhanced phosphate release with subsequent struvite production*. The extra feature of this K-struvite producing technology is that it extracts and recovers in one single operation two of the three main fertilizer compounds (NPK value). Potassium and Phosphorus are mined resources in contrast to N which as an unlimited source (e. g. atmospheric N_2).

5.3 Environmental performance and operational data

The struvite recovery rate depends on the process conditions and is determined by the concentrations of the parameters involved which determine the saturation ratio. The pH of the wastewater is a major process parameter. When conditions are correct the final chemical equilibrium results in residual PO_4 levels of 20-30 ppm $\text{PO}_4\text{-P}$. The initial P concentration in the incoming wastewater is 120-140 mg P/L. So, the recovery rate will be in the range of 75-85 %. The potassium concentration is ranging between 1000 – 1500 ppm and thus, potassium is not limiting. The decrease in the potassium concentration will hardly be noticed, because the PO_4 concentration is limiting the precipitation processes. Some additional Mg might be needed to achieve higher process yields. Caustic solution addition for pH adjustment can be estimated with 0.9 kg NaOH/ m^3 . As Mg demand, 0.2 kg MgCl_2/m^3 is estimated.

The reactor consists of an impeller mix reactor (11 kW impeller) with additional pumps (overall 15 kW) and a grit washer.

5.4 Cross-media effects

A number of wastewaters have relatively high in K and PO_4 contents, those can be used to recover the 2 key fertilizer ingredients. These types of wastewater result typically from food processing, manure treatment or from digestion processes such as digestate dewatering liquor. An important point of attention is the possible interference of NH_4 in terms of struvite formation. If pure K-struvite is required, NH_4 needs to be absent (e.g. wastewater from semi-conductor production) or needs to be removed first by a preceding treatment.

5.5 Technical considerations relevant to applicability (pretreatment, certain requirements)

This type of technology is only beneficial with initial PO_4 levels of 50-60 ppm $\text{PO}_4\text{-P}$ onwards in a main stream treatment. There is no specific pre-treatment necessary, if phosphorus is already present as PO_4 . Otherwise, an anaerobic preceding step ensures that condition, but simultaneously, NH_4 very likely results from that treatment step. Hence, for the formation of K-struvite, the ammonium has to be removed prior to its formation. Thus, the appropriate technology is an activated sludge type treatment. Of course, potassium needs to be available in a dissolved form as it usually is.

5.6 Economics

The amount of FeCl_3 saved can be estimated using the Fe/ $\text{PO}_4\text{-P}$ molar ratio of 1.5 (on 70% of the P load). This results in about 2000 kg of FeCl_3 or 1000 €/day. Also this amount of FeCl_3 added will result at least in a 1700 kg DM/day. Processing and disposal of the extra sludge are 75 €/ton or with 20% DM, this accounts up to 640 €/day. Hence, chemical savings correspond to cost savings of about 1640 €/day (0.5 – 0.6 million €/year). However, in addition, there will be other chemicals needed such as MgCl_2 and NaOH. Those cause additional costs of 250 €/day for MgCl_2 and 800 €/day for NaOH.

The struvite reactor causes the major costs and can be estimated for the size of 125 m^3/h with costs ranging from 0.75 to 1 million €.

5.7 Driving force for implementation

Besides the possibility of recovering phosphate/potassium and the production of a reusable fertilizer or fertilizer commodity product as well as the avoidance of disposing of phosphorus/potassium in a non-sustainable way, there are also other incentives to use this approach: reducing the salinity in the final effluent which renders its reuse as irrigation water more likely. This type of water supply will become more and more prominent in the coming years due to climatic changes and longer or more frequent drought phases.

5.8 Example plants

There is a K-struvite unit active on veal manure installed after the aerobic stage (Putten – Netherlands). Furthermore, a K-struvite production plant is commissioned in the frame of Circular Agronomics. More information is available on the Circular Agronomics homepage or in D3.3 (to be updated after M42 (July 2022), when D3.3 is available).