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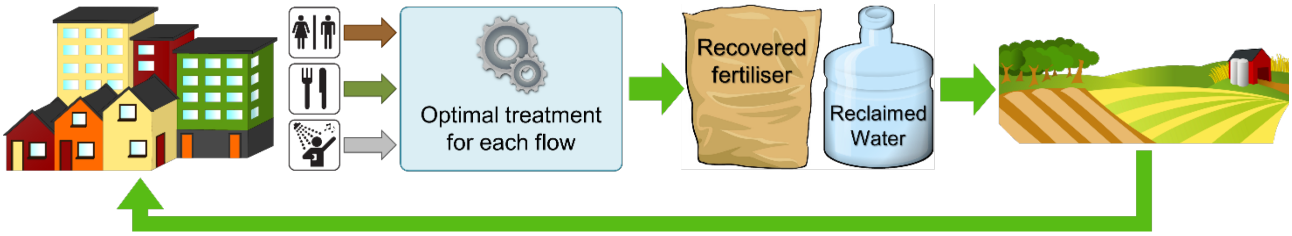
Integrated nutrient recovery from source-separated domestic wastewaters for application as fertilisers

Iemke Bisschops[a], Hamse Kjerstadius[b], Brendo Meulman[c], Miriam van Eekert[d]

[a] LeAF BV, P.O. Box 500, 6700 AM Wageningen, The Netherlands / *iemke.bisschops@wur.nl* (corresponding author)
[b] NSVA AB, Box 2022, 250 02 Helsingborg, Sweden / *hamse.kjerstadius@nsva.se*
[c] DeSaH BV, Pieter Zeemanstraat 6, 8606 JR Sneek, The Netherlands / *b.meulman@desah.nl*
[d] Wageningen University, Environmental Technology, P.O. Box 17, 6700 AA Wageningen, The Netherlands / *miriam.vaneekert@wur.nl*

Abstract

Source separation and decentralised treatment of domestic wastewaters for resource recovery have matured into a viable alternative for large-scale centralised treatment. The separate collection of toilet wastewater facilitates optimised treatment of the separate flows for efficient resource recovery.



Practical examples are set at the four demonstration sites of EU-project Run4Life[1]. Socio- economical and legislative aspects are important in the applicability of these concepts and recovered products, as well as hygienic safety, heavy metals and organic micropollutants. Depending on site- specific issues, different technologies can be integrated to recover products that meet the requirements of agriculture and society.

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[1] *Run4Life is the acronym of the project "Recovery and Utilization of Nutrients 4 Low Impact Fertiliser". It receives funding from the EU Horizon 2020 Research and Innovation programme under grant agreement number 730285.*

Introduction

Domestic wastewaters are carriers of organic matter and nutrients, largely originating from food: faeces, urine and food waste [1-3]. Throughout human history many different types of latrines, cesspits, toilets and sewers have been developed, abandoned and re-invented, with the earliest flush toilets dating back several millennia BC [4]. The fertiliser qualities of human waste were recognised and valued, with large amounts being collected in cesspits and bucket latrines, sold and applied in agriculture. This changed when the connection was made between excreta and the spreading of illness. It is now widely recognised that adequate sanitation is vital in preserving human health [5, 6]. Widespread sewer use started around the 1850's, and from then on it was slowly implemented in cities across the world, together with the use of flush toilets [4, 7]. The post-WWII rapid urbanisation period of 1950-1970 was accompanied by a large increase in the number of flush toilets and sewer connections, followed by the implementation of sewage treatment plants (STPs) [8].

Currently, over 90% of the urban population in Europe and Northern America has a sewer connection [9] and to protect not only human but also environmental health, STPs need to comply with stringent legislation for organic matter (OM) and nutrient removal. In the European Union, around 40% of the produced sewage sludge is used in agriculture [10, 11]. Agricultural use of sewage sludge is often limited by legislation regarding e.g. heavy metals and pathogens [12]. Emerging pollutants such as nanoparticles and pharmaceuticals and topics like antibiotic resistance are cause for debates on even stricter STP effluent limits and agricultural sludge use [13, 14]. At the same time the importance of wastewater and sludge as resource carriers in the circular economy is recognised [10, 14-16]. The production of artificial nitrogen fertilisers is highly energy consuming, and the easily accessible reserves of phosphorous rock are becoming depleted [17-20]. The recovery of nitrogen (N) and phosphorous (P) from domestic wastewaters is increasingly seen as a viable alternative.

During the last decades it has been demonstrated that resource recovery from domestic wastewaters is most efficiently done through source separation of toilet wastewater, thereby avoiding the dilution of organic matter and nutrients from excreta by mixing with flushing water and wastewaters from e.g. bathing and laundry washing. It is time for a paradigm shift in domestic wastewater management: optimal recovery of safe resources for agriculture, from source-separated domestic wastewater. In this review the recent developments in this field in Western Europe are highlighted.

Source separation and resource recovery in Western Europe

Source separation and decentralised treatment of domestic wastewaters for resource recovery started to gain interest in the 1990's, when several research groups questioned the standard practice of collecting mixed sewerage for removal of nutrients and organic matter and instead proposed a move towards resource recovery [21-23]. Urine diversion for use in agriculture [24-26], and vacuum toilets for collecting highly concentrated black water (BW) [27] followed by anaerobic digestion aiming to use the digestate in agriculture [28-30] were typical topics of research. Throughout the 2000's, different configurations of source-separated flows and recovery technologies were implemented on pilot and demonstration scale, including recovery of specific fertiliser products from digestate. Decentralised treatment and resource recovery is no longer only a part of the research domain: local governments, water authorities and companies are working together to initiate and implement the concept at larger scales. Examples are the currently implemented redevelopment areas H+ in Helsingborg, Jenfelder Au in Hamburg, Nieuwe Dokken in Ghent and Buiksloterham in Amsterdam. These projects each serve more than thousand people - a clear indication that the concept is past piloting and has reached maturity [31].

Four locations are currently further developed as full-scale demonstration sites in the EU H2020 project "Recovery and Utilization of Nutrients 4 Low Impact FErtilizer" (acronym: Run4Life), each with a different technological configuration. Resources are recovered from toilet wastewater (black water, BW), other domestic wastewaters (grey water, GW) and food waste (FW). The sites provide not only a test bed for selected technological innovations but will also be used for an in-depth analysis of social acceptance, governance models and legislation in different national contexts. Figure 1 provides an overview of the overall resource recovery concept, the different technologies implemented in Run4Life and the generated products.

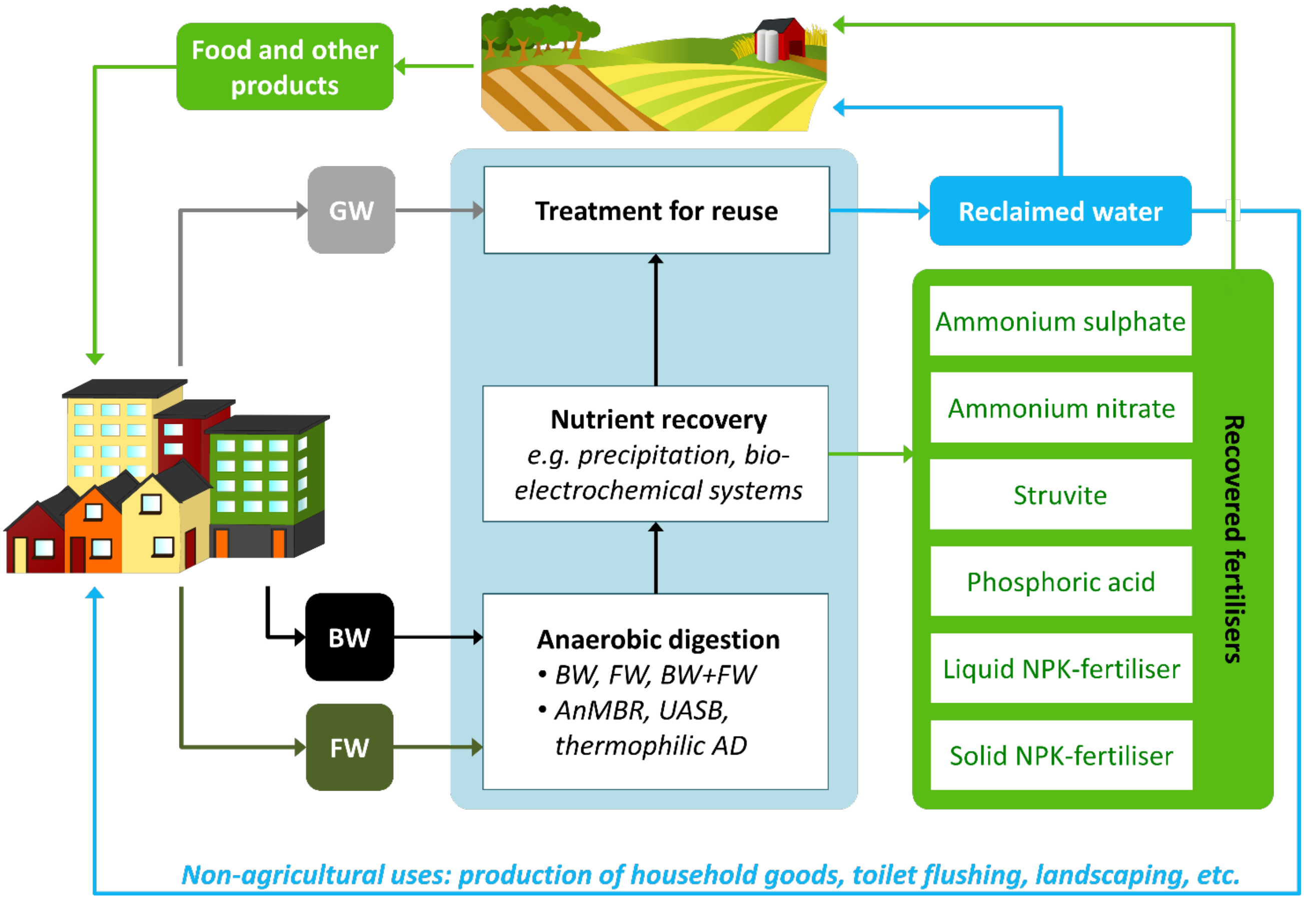


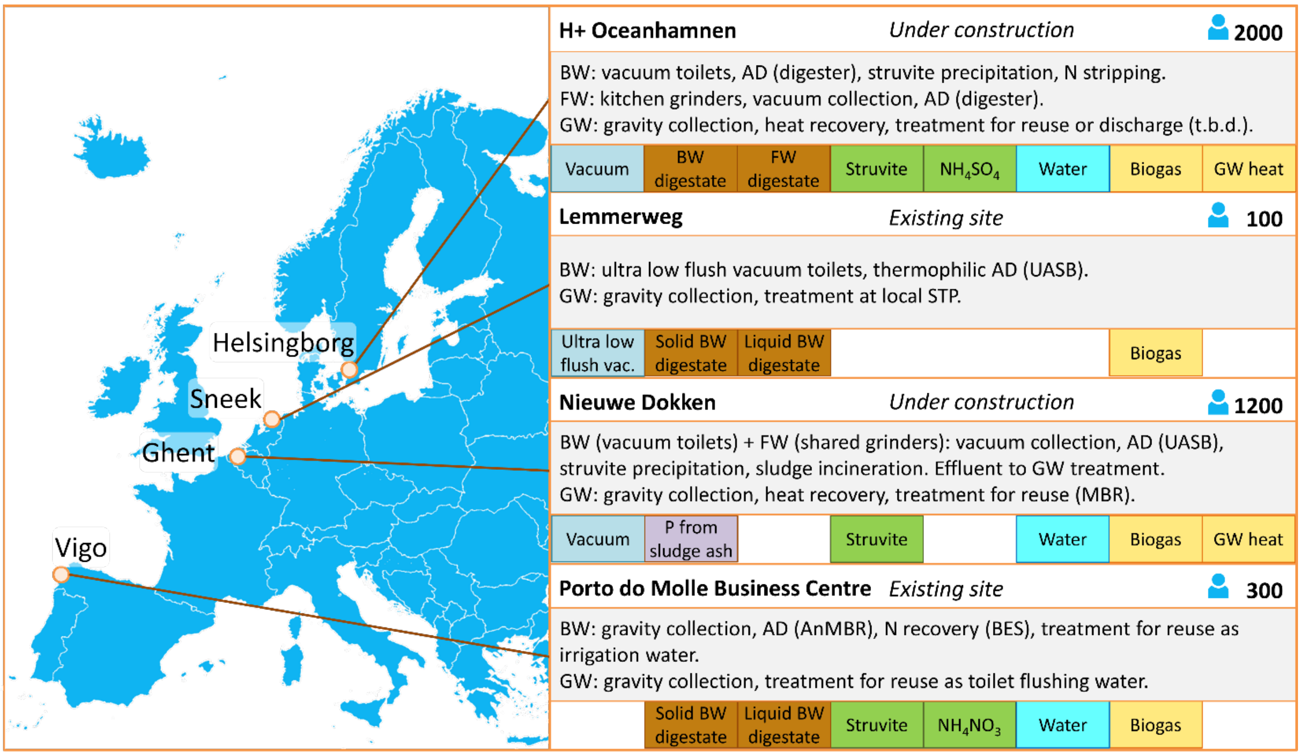
Figure 1. Technological resource recovery concept, focusing on water and nutrients. Energy recovery is not included. Scheme based on the technologies and products included in the H2020 project Run4Life. (BW: black water; FW: food waste; GW: grey water; AnMBR: anaerobic membrane bioreactor; UASB: upflow anaerobic sludge bed reactor; AD: anaerobic digestion; NPK: nitrogen, phosphorous, potassium).

Technologies and recovered resources

For resource recovery from source-separated domestic wastewaters many different technologies can be applied in different combinations [2, 32] to achieve optimal performance in a given setting. Which combinations of technologies are best suited in a certain situation depends on the local conditions. Anaerobic digestion (AD) is a core technology in decentralised resource recovery sanitation concepts [33, 34]. This is also clearly visible in Figure 2, an overview of the technologies and recovered products in the four demonstration sites of the Run4Life project. Each of the sites in Figure 2 includes AD as the first step for management of BW and/or FW, followed by nutrient recovery (especially P) and subsequent further treatment of the liquid phase.

From the basic concept of digestate use as organic fertiliser in the late 1990's there has been a development towards several options for recovery of separate N and P products, some of which are established (e.g. struvite precipitation) whereas other are currently at laboratory or pilot stage (e.g. bioelectrochemical systems, BES). By recovering separate products the market opportunities for recovered nutrients, as these can be mixed to obtain different desired fertiliser characteristics.

Currently, projects on nutrient recovery from domestic wastewaters are focussed on nitrogen and phosphorous. Also in the four sites that are part of the Run4Life project, potassium (K) is not recovered as a separate fertiliser compound. It will however be present in e.g. the liquid fertiliser produced by AD of the concentrated BW at the Lemmerweg site. At the moment only a few technologies are available to recover K from waste streams, e.g. electro dialysis, magnetic separation and adsorption [35] and due to the complicated organic matrix, those methods may have limited suitability for the domestic waste streams included in the project. Concentration by reversed osmosis and crystallisation as potassium struvite are viable options that have been demonstrated for a variety of waste streams, like manure and urine [35, 36].



H+ Oceanhamnen *Under construction* 2000

BW: vacuum toilets, AD (digester), struvite precipitation, N stripping.
FW: kitchen grinders, vacuum collection, AD (digester).
GW: gravity collection, heat recovery, treatment for reuse or discharge (t.b.d.).

Lemmerweg *Existing site* 100

BW: ultra low flush vacuum toilets, thermophilic AD (UASB).
GW: gravity collection, treatment at local STP.

Nieuwe Dokken *Under construction* 1200

BW (vacuum toilets) + FW (shared grinders): vacuum collection, AD (UASB), struvite precipitation, sludge incineration. Effluent to GW treatment.
GW: gravity collection, heat recovery, treatment for reuse (MBR).

Porto do Molle Business Centre *Existing site* 300

BW: gravity collection, AD (AnMBR), N recovery (BES), treatment for reuse as irrigation water.
GW: gravity collection, treatment for reuse as toilet flushing water.

H+ Oceanhamnen	Vacuum	BW digestate	FW digestate	Struvite	NH_4SO_4	Water	Biogas	GW heat
Lemmerweg	Ultra low flush vac.	Solid BW digestate	Liquid BW digestate				Biogas	
Nieuwe Dokken	Vacuum	P from sludge ash		Struvite		Water	Biogas	GW heat
Porto do Molle Business Centre		Solid BW digestate	Liquid BW digestate	Struvite	NH_4NO_3	Water	Biogas	

Figure 2. Full scale sites in the H2020 project Run4Life, the applied technologies and resulting products (image adapted from Run4Life communication and dissemination materials). Not all of the sites' technical features are included in the Run4Life activities.

Vacuum toilets and anaerobic digestion

The key function of AD is to convert organic matter into methane, while releasing phosphate and ammonia that become available for recovery. For efficient anaerobic digestion and subsequent nutrient recovery the input should be as concentrated as possible [30, 33, 34, 37]. Higher concentrations of biodegradable organics lead to higher biomass concentrations in the anaerobic reactors, and as a result the required reactor volume is smaller. Moreover, released nutrients (N and P) are more concentrated which allows recovery in higher concentrations. Concentrated BW can be achieved by using vacuum toilets. These use little water for flushing, commonly around 1-1.5 l/flush for conventional vacuum toilets. Within the framework of Run4Life, ultra-low dual flush vacuum toilets have been developed to provide an even more concentrated AD influent. These use 0.4-0.7 l/flush and will be implemented at the demonstration site in Sneek, The Netherlands (see also Figure 2).

Solid and liquid organic fertilisers

BW and FW digestate contain residual organic matter and a broad spectrum of macro and micro nutrients. Hygienisation is a prerequisite for the safe agricultural reuse of the anaerobic digestate and its solid or liquid fractions [12, 38]. A separate pasteurisation step can be avoided when the anaerobic treatment itself is carried out at thermophilic temperatures at appropriate minimum retention times [12, 39-42].

Phosphorous precipitation

Anaerobic digestate can be used for precipitation of struvite, a slow-release phosphate fertiliser. Struvite is currently produced at several decentralised and centralised STPs at a variety of scales [43, 44-45] and is probably one of the better known fertiliser products recovered from wastewater. Examples of alternative outlooks for phosphorous recovery through precipitation are the formation of calcium phosphate granules inside UASB reactors during anaerobic treatment of BW [46, 47] and electrochemical precipitation as struvite or calcium phosphates [48, 49].

Nitrogen stripping

Anaerobically digested BW contains around 1-1.5 g NH_4-N/litre [50] which can be recovered using conventional stripping methods and subsequent sorption in sulphuric or nitrous acid to form salts or solutions which are already in widespread agricultural use. Although energy and chemical intensive, stripping provides recycled products of high value to the agricultural market [51, 52]. Alternatively, ammonium may be recovered by applying biological electrochemical processes. These also involve stripping and sorption in an appropriate solution, but without the need of chemical dosing [53].

Energy

Domestic wastewater contains energy in the form of organic matter (BW and FW) and heat (GW). By optimally combining wastewater flows, treatment technologies and effluents, energy can be recovered and reused in the treatment system or elsewhere. Nearby sources of residual heat (e.g. factories) can also be included in the energy concepts [37].

Water

Another recoverable resource is water. Recovered water can be used as irrigation water in landscaping or agriculture, as process water in industry or as second-class water in households. Depending on the exact local requirements, additional treatment steps may be needed [32, 54].

Pathogens, heavy metals and organic micropollutants

For recovered resources to be used in food production, it is of vital importance that they meet the demands of the agricultural sector in terms of reliability, composition, quality and applicability. When recovering products from domestic wastewaters, hygienic aspects are a key aspect. Depending on the fertiliser produced this could be more or less of an issue. Mesophilic anaerobic digestion does not inactivate pathogenic activity or pathogens harbouring Antibiotic Gene Resistance (ARG), which is gaining more and more attention. For some pathogens inactivation is achieved under thermophilic anaerobic digestion [55]. The ammonium salts are generally produced after stripping of the nitrogen and subsequent sorption in an acid. The production process includes a phase separation between the source separated streams and the product. Nitrogen stripping and sorption is already applied in practice and such fertilisers have been admitted to the market [56]. For phosphate precipitates such as struvite this is different. These are in general formed in a reactor while still in contact with the treated source separated streams. In the process of validating these precipitates as safe fertilisers, research has been directed towards their hygienic qualities. A Dutch study showed that struvite produced from digested sludge at a municipal sewage treatment plant contained quite large amounts of other solids and was not hygienically safe [57]. However, after removal of the impurities the pathogen content of the struvite was comparable or lower than animal manure, which is normally applied as fertiliser.

In addition to hygienic safety, recovered fertilisers should be reliable with regard to the presence of organic micropollutants and heavy metals. The source of the recovered fertilisers will largely determine their heavy metal content. It has been demonstrated that their concentrations in source separated black water sludge are much lower than in sewage sludge, with only the exception of Zn, that was found in concentrations above the Dutch legal limit [58]. In several countries, sewage sludge is not allowed to be used in agriculture because of heavy metal concentrations that surpass the legal limits. This legislation also affects the use of sludge from anaerobically digested source separated black water, limiting the options for organic fertiliser recovery. Interestingly in view of the circular concept, heavy metals in source separated black water – including zinc - predominantly originate from dietary sources [58]. It has been shown that heavy metals are not incorporated in struvite crystals [59] and that struvite products contained only minor amounts of heavy metals and organic micropollutants, complying with Dutch legal limits [57].

As in mixed sewage, organic micropollutants (e.g. pharmaceuticals, personal care products, and others) are inevitably also present in most source separated waste streams. Being the main undiluted source of pharmaceutical residues, BW contains relatively high concentrations of these compounds. Concentrations of up to 100 times higher than in mixed sewage have been detected [60]. Depending on the nature of the compounds they may or may not be degraded in the recovery and treatment systems and/or incorporated in the fertilisers [61]. However, it was found that the uptake of organic pollutants in plants (wheat and carrot) was negligible when applying normal fertilisation practices. Calculations showed that several thousands of years of normal consumption were needed to reach the equivalent of one therapeutic daily dosage [60]. It should be noted that most research directed towards the fate of micropollutants in nutrient recovery systems has been done under mesophilic conditions. Anaerobic digestion under (hyper)thermophilic conditions in relation to micropollutants has not yet been subject of research. The fate of micropollutants under said conditions will be assessed within the framework of the Run4Life project.

Integrated implementation

In the successful implementation of the entire concept as visualised in Figure 1, many factors play a role. For recovered resources to be used in food production, it is of vital importance that they are hygienically safe. In addition, recovered fertilisers should meet the demands of the agricultural sector in terms of reliability, composition, quality and applicability. Technological challenges such as improved recovery rates and treatment efficiencies are frequent topics in wastewater literature, but socio-economic, legislative and practical issues are often decisive in social and market acceptance of source-separated sanitation concepts [31, 52, 62, 63-65, 66, 67]. All stakeholders should be taken into account when developing projects that include source-separated sanitation for resource recovery, to ensure acceptance of the systems and their products. Between countries, significant differences exist in environmental legislation and the acceptance of recovered resources, creating different playing fields for circularity [68]. Here, the Swedish certification system for black water fertiliser products [69] is a good example on how to achieve social acceptance. The BW certification system is in use to allow for the controlled and traceable reuse of treated BW and extracted products in agriculture, with the purpose to avoid health and environmental risks.

Research into the economic aspects of source separation and resource recovery systems has shown that comparing wastewater collection and treatment scenarios purely by economic factors is a complex matter, with not one system always performing better and a high dependence on local circumstances and the chosen system boundaries for comparison. In addition, to date, the current and future monetary and non-monetary value of resource recovery is insufficiently represented in cost-benefit calculations [62, 64, 65, 70].

Technological advances will be made in optimising the recovery methodologies, focussed on generating products that meet the reuse requirements of agriculture and society. Given the upcoming demand for K in fertiliser and the limited availability of potassium ores, the development of technologies for dedicated recovery of potassium is warranted. It is expected that these technologies will then be included in resource recovery sanitation concepts. Although usually at a slower pace, alongside the technological developments also policies are continuously reviewed and adapted, translating into changes in legislation. It is important that these transitions find an appropriate balance between precautionary actions to safeguard human and environmental health, and enabling the closing of cycles that is needed for a sustainable future.

Conclusions

Since their early development 30 years ago, modern source separated sanitation concepts have matured into reliable wastewater management and resource recovery systems: several European cities are currently building source separation systems on a previously unprecedented scale of several thousands of users. From the source-separated flows different macro nutrients (N, P, K) can efficiently be recovered for reuse as fertilisers, as well as energy and organic matter. The closing of nutrient cycles should be achieved while taking into account human and environmental health. Technological advances will lead to the production of recovered fertiliser products that are tailored to meet the reuse requirements of agriculture and society with respect to quality and safety. Pioneering projects such as those highlighted in this paper will generate vital knowledge to enable further advances. Local conditions should be taken into account when designing the systems, working in close cooperation with all relevant stakeholders as social and market acceptance of the technologies and recovered products is crucial for their successful implementation.

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