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The peer-reviewed published version of the paper contains an additional section on pathogens, heavy metals and organic micropollutants.

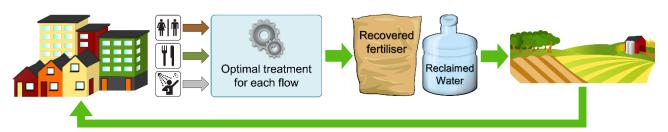
Integrated nutrient recovery from source-separated domestic wastewaters for application as fertilisers

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Abstract

During the last decades, source separation and decentralised treatment of domestic wastewaters for resource recovery has matured into a viable alternative for large-scale centralised treatment. The newest real estate development projects that integrate the concept are catering for thousands of people. Toilet wastewater is concentrated in a small volume, and collected separately from the other domestic wastewaters. This allows for optimal treatment of each wastewater type and offers better opportunities for efficient resource recovery than the centralised treatment of diluted mixed sewage.



Anaerobic digestion of toilet wastewater is a core technology, as it allows for energy recovery in the form of biogas while making N and P available for recovery. Socio-economical and legislative aspects play a key role in the applicability of these concepts and the recovered products. Depending on the local situation, different technologies can be applied to best fit the local requirements.

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].

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. 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. provides an overview of the overall resource recovery concept, the different technologies implemented in Run4Life and the generated products.

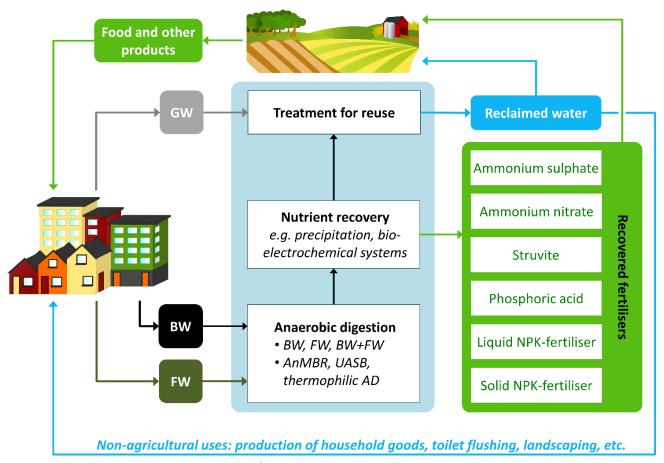


Figure 1. Technological resource recovery concept, focusing on water and nutrients. Energy recovery is not included. Scheme based on the technologies and products included in the H2020 project Run4Life. (BW: black water; FW: food waste; GW: grey water; AnMBR: anaerobic membrane bioreactor; UASB: upflow 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). 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 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 potassium from waste streams, e.g. electro dialysis, magnetic separation and adsorption [35] and due to the complicated organic matrix, those methods may only 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].

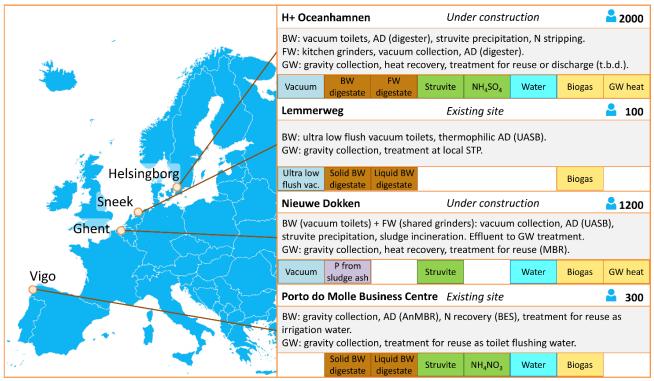


Figure 2. Full scale sites in the H2020 project Run4Life, the applied technologies and resulting products (image adapted from Run4Life communication and dissemination materials). Not all of the sites' technical features are included in the Run4Life activities.

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 (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 [43-45] 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 NH₄-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.

Integrated implementation

In the successful implementation of the entire concept as visualised in Figure 1, many factors play a role. 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, 54-59]. For recovered resources to be used in food production, it is of vital importance that they are hygienically safe. In addition, they should meet the demands of the agricultural sector in terms of reliability, composition, quality and applicability. 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. 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.

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 be recovered at an economically sustainable cost, together with other resources such as energy (biogas, heat) and water. Local conditions should be taken in to account when designing these systems, in close cooperation with all relevant stakeholders.

Acknowledgements

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