Phosphate and organic fertilizer recovery from black water

Taina Tervahauta

Thesis committee

Promotors
Prof. dr. ir. C.J.N. Buisman
Professor of Biological Recovery and Reuse Technology, Wageningen University
Prof. dr. ir. G. Zeeman
Personal chair in 'New Sanitation' at the Sub-department of Environmental Technology, Wageningen University

Co-promotor
Dr. ir. L. Hernández Leal
Scientific project manager
Wetsus, Centre of Excellence for Sustainable Water Technology

Other members
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Prof. dr. ir. L. Angenent, Cornell University, New York
Dr. ir. P. Van Der Maas, Waterlaboratorium Noord (WLN), Glimmen

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Taina Tervahauta

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Abstract

Soil degradation is one of the most crucial problems humanity is facing. This is induced by the use of artificial fertilizers that disregards the input of organic matter in soil. Moreover, phosphate fertilizers are predominantly sourced from phosphate rock – a resource that is declining in both quantity and quality. In this thesis the integration of treatment systems for black and grey water was investigated to improve resource recovery within source-separated sanitation concepts. Special focus was set on phosphate and organic fertilizer recovery from vacuum collected black water. Currently, the soil application of black water sludge is prohibited in the Netherlands due to elevated heavy metal concentrations. However, the use of the same guidelines for sewage and black water can be argued, since heavy metals in black water mainly originate from feces (52%–84%) while in sewage the contribution of feces is less than 10%. To distinguish these streams in the sludge reuse regulation, a control parameter can be implemented, such as the Hg and Pb content that is significantly higher in sewage sludge compared to black water sludge (from 50– to 200–fold). The heavy metals in feces and urine are primarily from dietary sources, and by promoting the black water sludge soil application over livestock manure and artificial fertilizers, the heavy metal content in the soil/food cycle could be further reduced. To improve energy recovery within source-separated sanitation concepts, the organic matter in grey water can be utilized by concentrating it in a bioflocculation unit. Anaerobic grey water sludge treatment with black water increased energy recovery by 23% in the upflow anaerobic sludge blanket (UASB) reactor compared to black water treatment, but introduced more heavy metals in the sludge, hindering its soil application. Instead, the flushing volume for black water vacuum collection could be lowered to decrease the energy consumption and heavy metal loading in black water treatment. A novel approach to simultaneously recover phosphate and energy was introduced by precipitation of calcium phosphate granules in anaerobic treatment of black water. Without any addition of chemicals, high purity calcium phosphate granules ($\geq$95%) were produced in the sludge bed of the UASB reactor. Further research is needed to render all of the phosphate in black water as pure calcium phosphate granules by optimizing the physical, chemical and biological parameters of the granulation process.

Keywords: black water, grey water, phosphate recovery, organic fertilizer recovery, anaerobic treatment, heavy metals

Contents

Chapter 1

General introduction

1.1 New paradigm for wastewater management

Humanity's growing hunger for consumption has resulted in vast environmental damage and depletion of natural resources. During the last decades, environmental protection has been riding on the concept of eco-efficiency to minimize material use and emissions. This has resulted in limited improvements in the state of the planet, and has prolonged the transition from a dysfunctional paradigm into a new way of thinking and doing. To move beyond eco-efficiency and zero emissions, the concept of eco-effectiveness has been adopted to describe the *cradle-to-cradle* design paradigm in production and consumption of goods and services. Within this concept the quality and productivity of materials is maintained or enhanced through their life cycle, known as *up-cycling* [21]. According to this concept, the reprocessing of biological materials is carried out by ecological processes, resulting in the regeneration and replenishment of natural systems. Urban water cycle is one of the key processes connecting human activity to natural systems. The health and well being of both human population and environment is therefore dependent on the integration of urban water systems with the natural systems. The method of *natural biological mineralization route* emphasizes anaerobic processes for the treatment of wastewater, combining the effectiveness of natural systems with the aim of resource recovery [107]. In order to facilitate this method, a new framework is required to replace the outdated, inflexible infrastructure of the current water systems, by strengthening the cooperation between different stakeholders [68, 72].

New sanitation with source-separation and on-site treatment of domestic wastewater is acknowledged as one of the most promising approaches to optimize resource recovery [103]. Within this approach, higher resource recovery efficiencies can be reached due to lower dilution, and higher quality recovery products can be attained due to separation of different waste streams. Domestic wastewater can be divided into *black water*, originating from toilet, and can be further divided into feces and urine using a urine diverting toilet, *grey water*, originating from shower, laundry and other washing activities, and *kitchen refuse* that is solid kitchen waste disposed by a kitchen grinder (Figure 1.1). Several different chemical, physical and biological treatment technologies, such as electrochemical and precipitation processes, membrane systems, and aerobic/anaerobic processes, have been introduced for resource recovery from the different domestic wastewater fractions [104]. Domestic wastewater holds a considerable energy potential [124], and new configurations on the existing treatment concepts have been proposed to improve energy recovery from source-separated waste streams [205]. Full scale results within *decentralized sanitation and reuse (DESAR)* concept on source-separation and on-site treatment of black water/feces and urine, grey water, and kitchen refuse have shown successful recovery of energy and nutrients, such as nitrogen and phosphorus [225]. The wide variety of technologies and treatment concepts for source-separated waste streams requires systematic scenario studies to realize the most optimal configuration for resource recovery at different locations and environmental conditions.

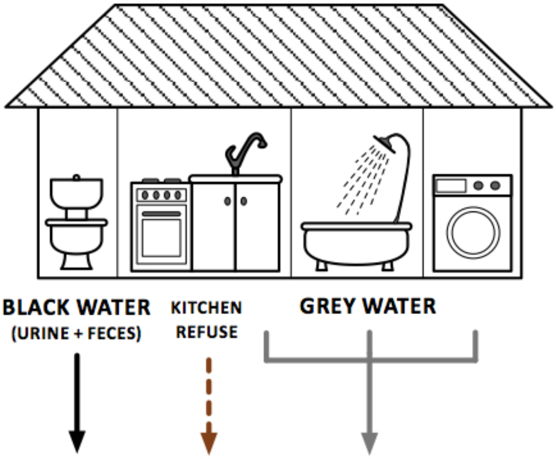

Figure 1.1: *Source-separated domestic wastewater streams*

1.2 Organic fertilizer recovery

Soil degradation through erosion and land use is one of the most crucial, yet largely hidden and underestimated problem humanity is facing. Rapid population growth, urbanisation and climate change induce out-of-balance carbon and nutrient flows, declining soil fertility at an alarming rate. Around 80% of the arable land in Africa is seriously degraded [197]. Several regions in Europe encounter decline in soil organic matter, and for example 25% of the soil organic carbon reserve in arable land in Belgium is lost during the past three decades [65]. The organic matter content of soil is a key factor determining its nutrient-holding capacity, and the supply of both organic and mineral nutrients is therefore essential to sustain food production [197]. Application of artificial fertilizers often dismisses the use of organic fertilizers to replace the outflow of carbon through harvested crops, breaking the balance between decomposition and assimilation of organic carbon in soil. The nature of the problem is insidious, as the rate of assimilation of stable organic carbon in soil is a very slow process compared to the rate of decomposition. To ensure global food security, soil fertility needs to be improved by recycling organic nutrients in agriculture.

A major portion of the nutrients leaving agriculture is in the food products. Since the human body excretes almost all the nutrients that are consumed with food, urine and feces contribute one of the largest fractions of the nitrogen and phosphorus flows in society [172]. In the past with cities close to the agricultural fields, human excreta, also known as 'night soil', was spread to the land to recycle back the nutrients (Figure 1.2 left side) [8]. However, as the cities grew, public health concerns influenced the sanitation revolution to move from a land-based system to a water-based system (Figure 1.2 right side). Currently, urine and feces are mixed with rest of the sewage, contaminating and diluting the valuable nutrients with industrial effluents, rain water and grey water, and either losing them during the treatment process or trapping them in polluted sewage sludge.

Figure 1.2: *Night soil collection in Delft (NL) in early 20th century (left) [113] and building of first sewer lines in Rotterdam (NL) in late 19th century (right) [112]*

In an urbanized society with a broken cycle of organic nutrients, other sources are used in agriculture to improve soil fertility. Peat is an ideal growing medium, used in horticulture and as a soil improvement in agriculture. In the Netherlands, 4.2 million m^3 of peat is imported yearly from Germany, Ireland and the Baltic states [18]. Peat extraction and transportation, however, induce soil degradation and increase CO_2 emissions, since carbon in the soil carbon stock is freed into the atmosphere as CO_2. A better alternative is to use locally produced bio-solids, such as organic municipal waste, by-products of agriculture and food industry, and domestic wastewater/black water sludge as organic fertilizers [172]. Livestock manure is conventionally applied in soil to improve the organic carbon and nutrient balance farm-on-site, yet the application of artificial fertilizers is often simultaneously practiced. To close the cycle of organic carbon and phosphorus between cities and agricultural land, the soil application of sewage sludge is promoted. However, the presence of heavy metals and other toxins in the sludge often restricts its use [168]. Within the European Union (EU), soil application of sewage sludge is regulated under the Directive 86/278/EEC that defines the maximum permissible heavy metal concentration and load in the sludge and soil [53]. Several member states, the Netherlands being among the most rigid, have further defined national limit values below the EU Directive [87]. For this reason the soil application of sewage sludge is prohibited in the Netherlands, and currently almost all of the sludge is incinerated, mineralizing the organic carbon into the atmosphere (Figure 1.3 left side) [17, 58]. By source-separation of black water, external input of heavy metals, such as industrial effluents, surface run-offs and grey water can be eliminated, and organic carbon and nutrients returned back to the soil/food cycle, reducing the need for peat and artificial fertilizers (Figure 1.3 right side). This is a particularly attractive alternative in organic farming, where the use of artificial fertilizers is prohibited.

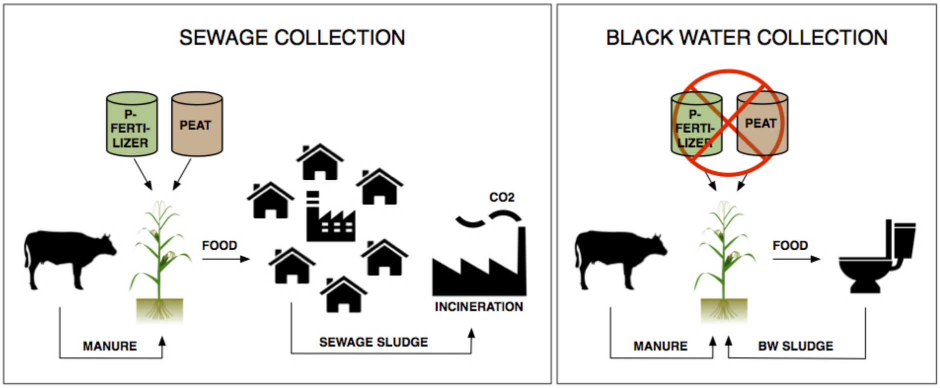

Figure 1.3: *Organic carbon and nutrient flows in sewage and black water collection*

Although the heavy metal content is significantly lower compared to sewage sludge, the soil application of black water sludge is prohibited in the Netherlands due to elevated Cu and Zn concentrations [41]. The use of the same guidelines for sewage and black water can be argued, since black water is mainly characterized by human originated content (feces and urine) while sewage is often a mixture of industrial and domestic wastewater, and rainwater. Furthermore, for example, by vacuum collection of black water, a minimum amount of flushing water is used to produce a concentrated stream with reduced input of heavy metals from tap water. To promote the recycling of organic carbon and nutrients, and to control the input of heavy metals into agricultural soil, a new targeted heavy metal regulation is required based on the origin of heavy metals in the waste stream. Additionally, the input of heavy metals from manure and artificial fertilizers into agricultural soil should be included in the regulation. In this way the recycling of organic fertilizers could be promoted while improving the soil quality by decreasing the heavy metal content in the soil/food cycle.

1.3 Phosphate recovery

Phosphorus is an essential and irreplaceable element in food production. To increase crop yield, external sources of phosphorus has been introduced in agriculture since mid 20th century [80]. Phosphate fertilizers are predominantly sourced from phosphate rock – a resource that is declining in both quantity and quality (Figure 1.4). Most of the remaining phosphate rock reserves are controlled by Morocco, China and US, leading to a geopolitically unstable situation in terms of phosphate rock supply and price [35]. Beside the political implications of phosphate rock mining, transporting and consumption of phosphate rock is highly inefficient. Significant amount of energy (20 MJ/kgP) is used for transporting of phosphate rock and phosphate fertilizers to end users [163], of which large part rely on fossil fuels. In terms of phosphate rock consumption, 25% from the 1 billion tonnes of phosphate mined over the past 50 years is wasted into water bodies and landfills [153]. Phosphate rock mining also involves significant amount of heavy metals and radioactive substances that are released into the environment, not only at the mining site, but also through the use of phosphate fertilizers. Phosphogypsum is a radioactive waste by-product produced five times the amount of phosphate mined, and needs to be stockpiled for disposal [193]. Uranium and cadmium (among other heavy metals) are geochemically associated with phosphate rock, and can be transferred into soil through the use of phosphate fertilizers [157]. To continue to meet the nutrient demand with phosphate rock mining, increased input of energy and resources are required together with increased volumes of waste and pollution. Therefore, from a political, economical and environmental point of view, phosphate recovery locally from waste streams is a prerequisite for global food security and sustainability.

Figure 1.4: *Phosphate rock mining in Togo, West Africa [28]*

Food production in Europe is mostly dependent on imported phosphorus [201]. In 2008 the net import of phosphorus in the Netherlands was 51 000 tonnes, while only 3% of the phosphorus in waste streams was recycled back to agriculture, the rest ending up in incineration ash or landfills [40]. Phosphorus in livestock manure accounts for up to 50% of the agricultural demand in Western Europe, but the spatial and temporal mismatch between supply and demand prevent the full utilization of this stream [165]. Human excreta accounts for 22% of the global annual phosphorus demand, and the sanitation infrastructure enables better match between collection, recovery and distribution of phosphorus compared to manure [165]. However, globally only 10% of the phosphorus in human excreta is recovered. Since human excreta are mixed with rest of the sewage in centralized collection systems, heavy metals and other toxins often prevent the utilization of the nutrients present in sewage sludge. Within EU only around 40% of sewage sludge is applied in soil, while the rest is either landfilled, incinerated or disposed in other way [87]. To increase phosphate recovery, several technologies have been introduced to produce a reusable phosphate product from various waste streams. The most common phosphate recovery technologies are precipitation of magnesium ammonium phosphate, known as struvite, and calcium phosphate from livestock manure and different fractions of domestic wastewater [105, 36].

Phosphate recovery from livestock manure, sewage and source-separated wastewater with different recovery products and technologies are presented in Table 1.1. In most of the recovery technologies, addition of either acid or base is required for selective precipitation of phosphate, and addition of either calcium or magnesium salt is required for increasing the supersaturation of the phosphate product. The technologies presented here have been studied on a lab– and pilot–scale, and few of them have also been established on an industrial scale with a commercialized phosphate recovery product, such as the Crystalactor®, PHOSNIX and OSTARA processes. However, the Crystalactor® process is currently out of operation due to high operation costs from chemical additions [85]. The recovery process, in particular the chemical additions influence greatly the feasibility of phosphate recovery. In addition, the wastewater source influences the feasibility of the recovery process and the purity of the product. For example, phosphate recovery from sewage sludge ash is energy intensive and costly, and involves high levels of heavy metals [111]. Other toxins present in wastewater, such as micropollutants, may also influence the quality of the recovered phosphate. Source-separation of domestic wastewater provides a stream with lower levels of heavy metals and micropollutants compared to sewage and manure [220]. Black water/feces and urine are therefore ideal sources for phosphate recovery.

Table 1.1: *Phosphate recovery from livestock manure, sewage and source-separated wastewater: recovery products and technologies*

Waste stream	P product	Recovery technology	Reference
Livestock manure			
Dairy manure	CaPhosphate	FBR	[70]
Swine manure	Struvite	FBR	[177]
Calf manure	Struvite	CSTR	[164]
Sewage			
Side stream	CaPhosphate	FBR (Crystalactor$^{®}$, NL)	[63]
of EBPR	CaPhosphate	FBR/Fixed bed/CSTR (P-ROC, Germany)	[12]
Membrane concentrate	Struvite	FBR	[20]
Sludge liquor	Struvite	FBR	[167, 22]
		(PHOSNIX, Japan)	[191]
		(OSTARA, Canada)	[146]
		(PRISA, Germany)	[144]
Digested sludge	Struvite	Acid extraction (SEABORNE, Germany)	[131, 222]
Sewage sludge ash	Al/CaPhosphate	Acid extraction (SEPHOS, Germany)	[158]
	Struvite	Thermochemical	[147, 176]
	CaPhosphate	Acid extraction	[142, 57]
Source-separated wastewater			
Urine	Struvite	CSTR	[219, 188]
		(SaNiPhos$^{®}$, NL)	[64]
		(SANIRESCH, Germany)	[156]
Black water	Struvite	CSTR	[94]

EBPR enhanced biological phosphorus removal
FBR fluidized bed reactor
CSTR continuously stirred tank reactor

Recovery of struvite is widely applied, mostly due to its spontaneous precipitation process and the co-precipitation of ammonium, resulting in a product that can be reused as a slow release fertilizer in agriculture [50]. However, recovered phosphate product needs to be certified in order to be used as a fertilizer, and the reuse of struvite in agriculture is therefore limited with the exception of few commercialized struvite products such as PHOSNIX and OSTARA. Moreover, struvite cannot be used as raw material in fertilizer industry using the existing process technology, due to the presence of ammonium and magnesium [51]. Recovery of calcium phosphate is more beneficial as it has the effective composition of phosphate rock, and can be therefore used as raw material in fertilizer industry. Calcium phosphate can also be used as a direct fertilizer, provided that the bioavailability of phosphate is sufficient. There is a need to recover phosphate as a high value product from a source-separated stream with reduced heavy metal content, in a process with minimum addition of chemicals.

1.4 Scope of this thesis

Source-separation of domestic wastewater presents a great potential for resource recovery. The aim of this project was to develop an integrated treatment system for black and grey water to improve resource recovery within source-separated sanitation concepts. Special focus was set on the recovery of phosphate and organic fertilizer from vacuum collected black water. Optimal resource recovery is dependent on the selection of existing treatment processes and the local environmental conditions. For this reason, the integration of different treatment processes at different levels of source-separation for energy, water and nutrient recovery is investigated in Chapter 2 using a mass and energy balance-based model. Heavy metal concentration of black water sludge is currently the most important parameter limiting its reuse in agriculture. In Chapter 3 the heavy metal mass balance of black water is determined and compared with sewage, and the primary origin of heavy metals is investigated to discuss the influence of black water sludge reuse on the heavy metal content in the soil/food cycle. Domestic wastewater holds a great energy potential, and the improvement of energy recovery is studied in Chapter 4 by anaerobic grey water sludge treatment with black water. Emphasis was also laid on the influence of grey water sludge addition on the excess sludge quality in terms of heavy metals to ensure its soil application. Recovery of a high value phosphate product, such as calcium phosphate, is a necessity for optimal phosphate recovery. In Chapter 5 a novel approach to phosphate recovery is presented by calcium phosphate granulation in anaerobic treatment of black water. Finally, a sanitation concept is proposed in Chapter 6 based on the knowledge gathered in this thesis, and the outline of the following research is further discussed.

Chapter 2

Prospects of source-separated sanitation concepts: A model-based study

Abstract

Separation of different domestic wastewater streams and targeted on-site treatment for resource recovery has been recognized as one of the most promising sanitation concepts to re-establish the balance in carbon, nutrient and water cycles. In this study a model was developed based on literature data to compare energy and water balance, nutrient recovery, chemical use, effluent quality and land area requirement in four different sanitation concepts: (1) centralized; (2) centralized with source-separation of urine; (3) source-separation of black water, kitchen refuse and grey water; and (4) source-separation of urine, feces, kitchen refuse and grey water. The highest primary energy consumption of 914 MJ/capita(cap)/year was attained within the centralized sanitation concept, and the lowest primary energy consumption of 437 MJ/cap/year was attained within source-separation of urine, feces, kitchen refuse and grey water. Grey water bioflocculation and subsequent grey water sludge co-digestion decreased the primary energy consumption, but was not energetically favorable to couple with grey water effluent reuse. Source-separation of urine improved the energy balance, nutrient recovery and effluent quality, but required larger land area and higher chemical use in the centralized concept.

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2.1 Introduction

Separation of different domestic wastewater streams and targeted on-site treatment of these streams for resource recovery has been recognized as one of the most promising concepts to re-establish the balance in carbon, nutrient and water cycles [103, 141, 218, 227]. Domestic wastewater can be divided into two major streams: concentrated stream of black water (feces and urine) and kitchen refuse, and less concentrated stream of grey water from washing activities, such as laundry, shower and bath. Black water can be further divided into urine and feces using urine diverting toilets or urinals. Energy and nutrients can be recovered primarily from the concentrated streams, while the less concentrated stream serves as an alternative water source.

Key technology for energy recovery from source-separated streams is anaerobic treatment of black water or feces and kitchen refuse in an upflow anaerobic sludge blanket (UASB) reactor [227, 91]. Nutrient recovery and pollutant removal from the UASB reactor effluent can be established by struvite precipitation, autotrophic nitrogen removal using oxygen limited anaerobic nitrification denitrification (OLAND) reactor and a post-treatment, such as a trickling filter (TF), to remove remaining organic material [227]. Due to operational conditions, such as a lower buffer capacity of the OLAND reactor effluent compared to the UASB reactor effluent, the struvite precipitation is preferred after the nitrogen removal [212].

Urine separation can be employed in two different approaches: in the source-separation-based sanitation and coupled with the existing centralized sanitation. Separation and direct reuse of urine on agricultural land can be used to increase nutrient recovery, improve wastewater effluent quality and to decrease operational energy consumption, due to lower nutrient concentrations in wastewater [122]. However, collection and reuse of source-separated waste streams, urine in particular, also involves social and cultural issues requiring attention when implementing new technology [101].

Commonly used treatment systems to remove organic material and nutrients from grey water include sequencing batch reactor (SBR) [75] and constructed wetlands (CW) [9]. Due to the considerably high land area requirement, the use of CW is not suitable for densely populated areas, such as the Netherlands [23]. One option could be, however, to implement CW as a green roof [9]. To utilize the organic material present in grey water, excess sludge from the grey water treatment system can be potentially co-digested in the UASB reactor instead of using energy-intensive sludge transport and disposal [74]. However, the possible inhibitory effect of surfactants present in grey water sludge on anaerobic digestion should be investigated [60]. To avoid extensive mineralization of grey water sludge, a bioflocculation unit, such as a high loaded membrane bioreactor (MBR) or A-trap from the AB-process [16], can be used to concentrate grey water at short hydraulic and sludge retention times (HRT and SRT). A post-treatment system (such as TF) can be applied to remove the remaining organic material from grey water effluent prior to reuse.

Quantitative tools, such as Material Intensity per Service unit (MIPS), exergy analysis and Life Cycle Assessment (LCA) have been used to draw energy and material balances of different centralized and source-separation-based sanitation concepts [140, 71, 11]. These studies present data on energy consumption and production, material intensity, and emissions of source-separated feces, urine and grey water treatment and centralized wastewater treatment with and without urine separation. For more in depth insight into the urban water cycle, Makropoulos et al. [118] developed an Excel/Matlab-based decision support tool for sustainable integrated urban water management, including domestic wastewater streams and rain water. Extensive information was provided on different household components for water use and options for water treatment and reuse, producing a complete water balance. A study on economic viability and critical influencing factors of different implementation scales of black water and grey water source-separation compared to the centralized sanitation was conducted by Thibodeau et al. [186]. van Beuzekom et al. [198] conducted a social cost-benefit analysis on different sanitation concepts in Geerpark Heusden, a neighborhood in the Netherlands. This study compared centralized sanitation with different levels of source-separation of wastewater and different scales for the treatment of source-separated wastewater in terms of livability, safety, health, biodiversity and affordability. No studies, however, have investigated the influence of urine separation combined with different grey water treatment configurations and grey water sludge co-digestion on the energy and material balances of the sanitation concepts. The objective of this study was to present energy and water balances, nutrient recovery, chemical use, effluent quality and land area requirement of the centralized and source-separation-based sanitation concepts with and without urine separation, and with different configurations of grey water treatment.

2.2 Materials and Methods

2.2.1 Construction of the model

An Excel based model was developed based on literature data for the comparison of 4 sanitation concepts: (1) centralized sanitation, (2) centralized sanitation with source-separation of urine, (3) source-separation of black water, kitchen refuse and grey water, and (4) source-separation of urine, feces, kitchen refuse and grey water (Figure 2.1), from which concept 1 is applied on a full scale and concepts 2, 3 and 4 are demonstrated on a pilot or lab scale. These concepts were compared in terms of energy consumption and production, water saving and reuse, nutrient recovery, chemical use, effluent quality and land area requirement. The energy and material balances were based on collection, transport and treatment of wastewater leaving out the energy and materials used in the construction and maintenance of the required infrastructure. The model was tailored for European circumstances with a specific focus on the Netherlands. However, with small modifications on data input the model is applicable also in other circumstances.

The model was constructed from *location specific data* on environmental temperature, tap water temperature and distances to a sewage sludge incineration plant and agricultural land, *general data* on water consumption of different appliances and wastewater characteristics, and *treatment system specific data* on operational conditions, reactor performance, sludge production, energy consumption and energy production. The energy and water balance, recovered nutrients, chemicals used, effluent quality and land area requirement for each treatment system was then calculated using energy and mass balances based on the selected data.

2.2.2 Data inventory: Location specific data

Wastewater in concepts 1 and 2 were considered to be treated centralized (10 000 or more people), and the urine collection (concepts 2 and 4) and the treatment of black water or feces, kitchen refuse and grey water were considered to be community-on-site (100–10 000 people). Average environmental temperature of 10°C [89] and tap water temperature of 12°C [217] of the Netherlands were used. The distance from the centralized wastewater treatment plant to the sewage sludge incineration plant was set to 10 km [217] and the distance from the on-site collection to agricultural land

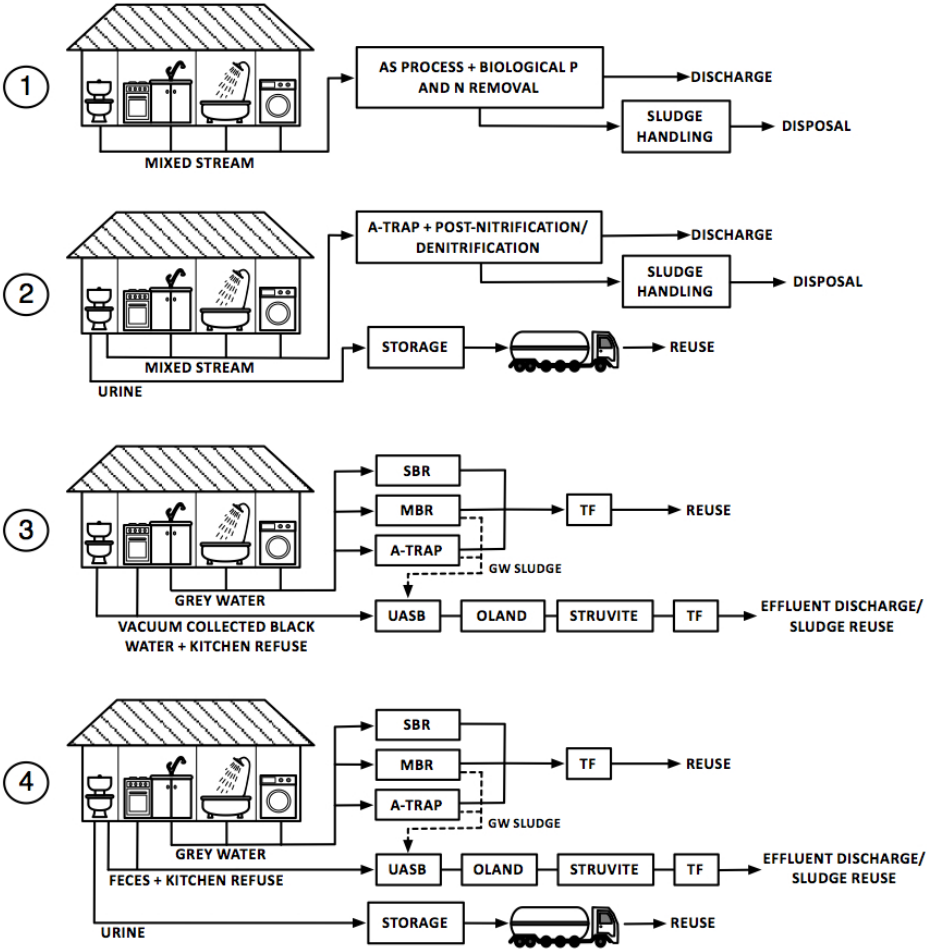

Figure 2.1: *Sanitation concepts included in the model with wastewater streams and corresponding treatment systems (AS = activated sludge process, SBR = sequencing batch reactor, MBR = membrane bioreactor, A-trap = A-stage of AB-process, TF = trickling filter, UASB = upflow anaerobic sludge blanket reactor, OLAND = oxygen limited anaerobic nitrification denitrification)*

was assumed to be 50 km, as a typical distance in the Netherlands. The influence of the transport distance on feasibility of the sanitation concepts was further discussed in the sensitivity analysis.

2.2.3 Data inventory: General data

The toilet type selected for concept 1 was a normal flush toilet, for concept 2 a urine diverting toilet (gravity), for concept 3 a vacuum toilet and for concept 4 a urine diverting toilet (gravity/vacuum). The water consumption of different toilets and kitchen grinder is presented in Table 2.1.

Table 2.1: *Water consumption of different toilets and kitchen grinder*

Parameter	Unit	Water use
Normal flush toilet (concept 1)	L/cap/d	34^{1}
Vacuum toilet (concept 3)	L/cap/d	6^{2*}
Urine diverting toilet (gravity) (concepts 2 and 4)	L/cap/d	5^{3*}
Urine diverting toilet (vacuum) (concept 4)	L/cap/d	2^{**}
Kitchen grinder (concepts 1, 2, 3 and 4)	L/cap/d	0.6^{2}

(1) [27]
(2) [92]
(3) [102] (0.2 L for urine and 4 L (assumed) for feces per flush)
*based on production of 1 time feces and 5 times urine per day
**based on 0.2 L for urine [102] and 1 L for feces per flush [92]

As a common practice in the centralized approach, the wastewater influent in concepts 1 and 2 was considered to consist of domestic wastewater, rain water runoffs and some industrial effluents ending up with a daily flow of 300 L/cap [217]. For better comparison between centralized and source-separation based sanitation concepts, the pollutant loading in the wastewater influent was considered to originate only from the domestic wastewater streams of urine, feces, kitchen refuse and grey water, and sludge rejection water from sludge dewatering, forming a daily loading of 176 gCOD/cap, 21 gTN/cap and 3.6 gTP/cap (concept 1), similar to the study of Wilsenach and van Loosdrecht (2006) [217]. Although kitchen refuse was not included in the study of Wilsenach and van Loosdrecht (2006) [217], the pollutant loading from kitchen refuse was considered to replace the pollutant loading from industrial effluents in this study.

Table 2.2 presents the characteristics of different domestic wastewater streams. In every sanitation concept the pollutant loading in the wastewater influent was calculated as a sum of the according sub-streams, and in concepts 3 and 4 the daily flow was calculated as a sum of the pollutant loading and the water consumption of the toilet and the kitchen grinder.

Table 2.2: *Domestic wastewater characteristics*

Parameter	Unit	Feces	Urine	Kitchen refuse	Grey water
Temperature	°C	37*	37*	20*	32^2
Volume	L/cap/d	0.1^1	1.4^1	0.2^1	79^4
COD	g/cap/d	50^1	11^1	59^1	52^1
BOD_5	g/cap/d	24^1	5.5^1	37**	27^1
TSS	g/cap/d	30^1	40^1	79^1	55^1
TN	g/cap/d	1.8^1	9^1	1.7^1	1.2^1
NH_4^+–N	g/cap/d	1.2^3	9^5	-	0.1***
TP	g/cap/d	0.5^1	0.8^1	0.2^1	0.4^1
PO_4^{3-}–P	g/cap/d	0.2^3	0.3^3	-	0.1***
K	g/cap/d	0.9^1	2.8^1	0.2^1	0.8^1

(1) [92]
(2) [76]
(3) [42] (NH_4^+–N/TN ratio of 0.7)
(4) [27]
(5) [217] (TN=NH_4^+–N in urine)
* based on body temperature (feces and urine) and average room temperature
** based on COD/BOD ratio of 1.6 [181]
*** based on NH_4^+–N/TN ratio of 0.1 and PO_4^{3-}–P/TP ratio of 0.35 [76]

2.2.4 Data inventory: Treatment system specific data

The wastewater treatment system in concept 1 was based on an activated sludge process (AS process) with biological phosphate and nitrogen removal, and in concept 2 on an A-trap (A-stage of AB-process [16]) with a post-nitrification/denitrification step according to the study of Wilsenach and van Loosdrecht (2006) [217]. As the wastewater in concept 2 was without the input of urine, a high loaded process with a short SRT and a post-treatment step was assumed to be sufficient for pollutant removal. Urine in concepts 2 and 4 was considered to be collected on-site with a collection degree of 75% [101], first stored for 6 months on-site and then transported to agricultural land to be used as a fertilizer by spreading. As a result of the breakdown of urea during storage, the high ammonium content and the increased pH ensures the hygienization of urine [207], and is recommended by the World Health Organization (WHO) for safe use of urine in agriculture [215]. The risk of ammonia emissions is prevented by using non-ventilated storage and handling. The treatment systems applied for black water or feces, kitchen refuse and grey water in concepts 3 and 4 are presented in Figure 2.1. Table 2.3 presents the pollutant removal efficiencies of the different treatment systems. The removal efficiencies in the AS process in concept 1 were according to existing wastewater treatment plants in the Netherlands.

Table 2.3: *Pollutant removal efficiencies (%) of biological reactors in concept 1 and 2, and of Upflow Anaerobic Sludge Blanket reactor (UASB), Oxygen Limited Anaerobic Nitrification Denitrification (OLAND), Struvite precipitator, Trickling Filter (TF), Sequencing Batch Reactor (SBR), A-stage of AB-process (A-trap) and Membrane Bio Reactor (MBR) in concepts 3 and 4*

			Concepts 3 and 4							
Parameter	Concept 1	Concept 2	Black water/feces and kitchen refuse					Grey water		
			UASB	OLAND	STRUVITE	TF	TOTAL	SBR	A-TRAP	MBR
COD	92^1	92^7	83^2	53^2	-	85^3	99^{**}	90^4	42^5	75^6
BOD_5	98^1	92^*	83^2	53^*	-	85^3	99^{**}	90^*	42^*	75^6
TSS	95^1	92^*	83^2	-	-	85^3	97^{**}	76^4	42^*	$\geq 95^6$
TN	80^1	72^7	1^2	73^2	9^8	-	76^{**}	35^4	36^5	81^6
TP	82^1	79^7	33^2	-	96^8	-	98^{**}	28^4	40^5	65^6

(1) [151]
(2) [94]
(3) [181] (based on standard rate filter with hydraulic loading of 1-4 m^3/m^2*d)
(4) [75]
(5) [45]
(6) [74]
(7) [217]
(8) [43]
*assumed based on COD removal
**calculated as total removal efficiency of UASB, OLAND, Struvite and TF

Incineration was selected for excess sludge treatment in concepts 1 and 2, as it is the most common practice in the Netherlands [58]. Complete sludge treatment consisted of anaerobic digestion to produce methane, sludge dewatering, transport of dewatered sludge to an incineration plant and sludge incineration. Sludge rejection water from sludge dewatering was recycled back to the influent. Excess sludge from the UASB reactor and the SBR (concepts 3 and 4) was considered to be transported to agricultural land for spreading without dewatering.

2.2.5 Calculations for energy balance

The total primary energy consumption in the sanitation concepts was calculated according to Eq. 2.1

$$E_{total} = E_{collection} + E_{treatment} + E_{urine/sludge\ transport} - E_{methane} \tag{2.1}$$

where $E_{collection}$ was the energy requirement for the collection and transport of wastewater, $E_{treatment}$ was the energy requirement for all the biological, chemical and physical treatment units for mixed wastewater stream, excess sludge and source-separated urine, black water/feces, kitchen refuse and grey water, $E_{urine/sludge\ transport}$ was the energy requirement for urine and excess sludge transport, and $E_{methane}$ was the energy production as methane. All the energy parameters were calculated as primary energy by converting the electrical energy (collection, aeration, mixing and pumping) using efficiency of 0.31 based on the European electricity mix [190].

$E_{collection}$ was the energy requirement for the gravity sewers with lifting stations (20 kWh/cap/y) [199] in concepts 1 and 2, for the vacuum collection and transport of black water and kitchen refuse (25 kWh/cap/y) [227] in concept 3, and for the vacuum collection and transport of feces and kitchen refuse (8 kWh/cap/y) in concept 4 (assumed to be $\frac{1}{3}$ of the energy requirement for the black water vacuum collection according to the water consumption ratio of $\frac{2}{6}$ L). Urine separation in concept 2 was assumed not to have a significant effect on the total wastewater flow and thus, on the energy requirement for the collection. Due to short wastewater transport distances in semi-centralized sanitation, the energy requirement for the gravity urine diverting toilet was assumed to be insignificant. The collection also included the energy consumption for the kitchen grinder (5 kWh/cap/y) [227] in all of the sanitation concepts.

In concepts 1 and 2 $E_{treatment}$ consisted of the following energy parameters. $E_{aeration}$ was the aeration energy required to oxidize organic matter and nitrogen in the AS process, A-trap and post-nitrification step, and was calculated based on an energy requirement of 2.2 MJ/kg$COD_{converted}$ and 14 MJ/kg$N_{converted}$ [122]. The aeration energy was calculated based on the fraction of oxidized COD of the total COD removed (43% in concept 1 and 22% in concept 2), and the fraction of nitrified N of the total N removed (94% in concept 1 and 76% in concept 2) [217]. E_{mixing} was the energy requirement for mixing of the biological reactors and the anaerobic digester, and $E_{pumping}$ was the energy requirement for pumping of the internal flows, return activated sludge and excess sludge to the anaerobic digester [217]. In concept 1 additional mixing energy of 5 MJ/kg$P_{removed}$ originated from the biological phosphorus removal [122]. $E_{heating(digester)}$ was the energy required to heat up the influent (excess sludge) to the operational temperature of the digester and to compensate heat loss through the digester walls. The primary energy required to heat up the influent was calculated according to Eq. 2.2

$$\Delta Q = m * C * \Delta T \tag{2.2}$$

where ΔQ is the required energy (J), m is the mass of liquid (g), C is the specific heat capacity of water (4.2 J/g°C) and ΔT is the temperature difference between the influent temperature and the operational temperature of the reactor. The influent temperature of the digester (concept 1 and 2) was considered to be the tap water temperature (12°C). The primary energy required to compensate heat loss was calculated according to Fourier's law presented in Eq. 2.3

$$E_{heat} = \Phi = -\lambda * A * \frac{dT}{dx} \tag{2.3}$$

where Φ is the heat transfer (W), λ is the thermal conductivity of the isolation material (W/m*k), A is the heat transfer area, dT is the temperature difference across the isolation material (K) and dx is the thickness of the isolation material (m). Mineral wool with thermal conductivity of 0.04 W/m*k and thickness of 0.05 m was considered to be used as isolation material [91]. The area of heat transfer was considered to be the surface area of the reactor (calculated from the volume and dimensions of the reactor presented under the sub chapter *Calculations for reactor dimensions and land area requirement*) and the temperature difference was considered to be the difference between the environmental temperature (10°C) and the operational temperature of the reactor (35°C). $E_{dewatering}$ and $E_{incineration}$ were the primary energy requirements for dewatering of the digested sludge and for incinerating the dewatered sludge according to the study of Wilsenach and van Loosdrecht (2006) [217], from which they were recalculated to primary energy using efficiency of 0.31. The heat production in the incineration of sludge was taken into account in the energy requirement.

$E_{sludge\ transport}$ was the energy requirement for transporting of dewatered sludge to the incineration plant and was calculated based on a primary energy requirement of 4.8 MJ/ton/km (including empty return trip) [217]. $E_{urine\ transport}$ was the energy requirement for transporting of urine from the on-site collection to agricultural land, and was calculated based on the energy requirement of transporting described above.

$E_{methane}$ was the energy produced as methane in the digestion of excess sludge and was calculated by taking into account the different excess sludge compositions in concepts 1 and 2, originating from the different SRTs (12 d and 0.8 d, respectively). As presented in the study of Wilsenach and van Loosdrecht (2006) [217], excess sludge from the A-trap was considered to consist of 25% adsorbed substrate and 75% biomass. The methanization level of the adsorbed substrate was assumed to be 73% [52]. No adsorbed substrate was considered in concept 1 due to the high SRT. The fraction of biodegradable biomass in concept 1 was assumed to be 45% and in concept 2 65%, and the methanization level of this fraction was considered to be 90% [217]. The volume of the produced methane was calculated using a theoretical methane production of 0.35 L/gCOD$_{converted}$ and the primary energy production from methane was calculated using the volume of methane and the calorific value of methane (35.8 MJ/m^3) [181].

The sludge production in the AS process (concept 1) and the A-trap (concept 2) was calculated according to Tchobanoglous et al. (2004) [181] (Eq. 2.4)

$$P = Y * Q * (S_0 - S) \tag{2.4}$$

where P is the sludge production (kgVSS/d), Y is the sludge yield (kgVSS/kgBOD$_{removed}$), Q is the influent flow (m^3/d), S_0 is the influent BOD concentration (mg/L) and S is the effluent BOD concentration (mg/L). Sludge yield of 0.58 kgVSS/kgBOD$_{removed}$ was used for the AS process (SRT 12d) and 0.85 kgVSS/kgBOD$_{removed}$ for the A-trap (SRT 0.8d) at 12°C. The sludge production as total solids was calculated using a VSS/TSS ratio of 0.85 [181]. The total wet sludge production was calculated using a dry solid content of 2.5% and the total dry sludge production (after dewatering) was calculated using a dry solid content of 20% [217]. In concept 1 additional sludge production of 3.3 kgTSS/kgP$_{removed}$ was assumed to originate from the biological phosphorus removal [122].

The composition of the sludge rejection water (COD, TN and TP) was defined as the difference between the digester influent (excess sludge from the AS process and A-trap) and the COD converted into methane and nitrogen and phosphorus incorporated into the anaerobic biomass. The amount of biomass produced in the digester was calculated using a biomass yield of 0.08 gVSS/gCOD$_{converted}$, and the amount of nitrogen and phosphorus incorporated into the biomass was calculated using fractions of 0.12 gN/gVSS and 0.03 gP/gVSS, respectively [181]. All of the nitrogen and phosphorus in the sludge rejection water was considered to be in the inorganic form of NH_4^+ and PO_4^{3-}.

In concepts 3 and 4 E$_{treatment}$ consisted of the following energy parameters. E$_{heating(UASB)}$ was the energy required to heat up the influent to the operational temperature of the reactor and to compensate heat loss through the reactor walls, calculated as described above with the digester in concepts 1 and 2. The influent temperature of the UASB reactor was calculated from the mass proportions of the according wastewater sub-streams (Table 2.2). In the case of grey water sludge co-digestion in the UASB reactor, the influent temperature was adjusted with the temperature of grey water sludge that was assumed to be the environmental temperature (10°C). No heating energy for other treatment steps were taken into account. E$_{OLAND}$ was the energy requirement for the OLAND reactor and was derived from the rotating power requirement of the rotating biological contactor according to Fujie et al. (1983) [59] (Eq. 2.5).

$$P(w) = \lambda_1 * N^2 * D^2 * A \tag{2.5}$$

where A is the surface area of discs (m^2), λ_1 is the frictional constant ($8.6*10^{-6}$ kWmin^2/min^4), N is the rotational speed of a disc (min^{-1}) and D is the disc diameter (m). The surface area of the discs was calculated from the total nitrogen load and the biofilm load (6300 mgN/m^2/d [126]). The disc rotational speed of 3 min^{-1} [212] and the disc diameter of 1 m [59] were selected. E$_{Struvite}$ was the energy requirement for the struvite precipitation and was calculated based on an electricity consumption of 3.8 kWh/kg$N_{influent}$ [126]. E_{TF} was the energy requirement for the trickling filter as a post-treatment step in both black water and grey water treatment lines and was calculated based on an average electricity consumption of 3 kW/1000 $m^3{}_{influent}$ [181]. E_{MBR} was the energy requirement for the MBR and was calculated based on an average electricity consumption of 0.3 kWh/$m^3{}_{greywater}$ [125]. The electricity consumption for the OLAND reactor, struvite precipitator, TF and the MBR was converted to primary energy using efficiency of 0.31 [190]. E_{SBR} and E_{A-trap} were the energy requirements for grey water treatment in the SBR and the A-trap, respectively, consisting of energy consumption for pumping and aeration. Energy consumption for pumping was calculated with Eq. 2.6 according to Karassik et al. (2001) [86]

$$E_{pump}(kW) = \frac{Q(m^3/d) * H(m) * specific\ gravity\ of\ fluid}{367.7 * \eta} \tag{2.6}$$

where Q is the flow rate, H is the pump head and η is the pump efficiency. For the SBR the pump head was considered to be the height of wastewater in the reactor. For the A-trap the pump head was considered to be the height of the buffer tank for influent pump and the height of the aerated grit chamber and settling tank for the two intermediate pumps (calculations for pump head are presented in the sub chapter *Calculations for reactor dimensions and land area requirement*). The specific gravity of fluid was considered to be 1 and η was set to 0.68 according to the study of Wilsenach and van Loosdrecht (2006) [217]. The total energy consumption for pumping in the SBR was calculated from the energy consumption for two pumps: influent and effluent pump, feeding and discharge time of 15 min each and a total cycle time of 360 min [75]. The total energy consumption for pumping in the A-trap was calculated by assuming the pumping to be continuous. The energy requirement for pumping of the UASB influent was calculated to be insignificant and was not included in the energy balance. The energy consumption for aeration in the SBR and the A-trap was calculated according to the energy requirement of 2.2 MJ/kg$COD_{converted}$ [122]. The amount of oxidized COD in the SBR was calculated by defining the total amount of biodegradable COD removed in the reactor using a $COD_{biodegradable}/BOD_5$ ratio of 1.6 g/g [181] and excluding the amount of COD removed in the sludge using a sludge yield of 0.12 kgVSS/kgCOD [75] and a COD/VSS ratio of 1.4. The amount of oxidized COD in the A-trap was assumed to be 11% of the incoming COD [45].

Nitrogen removal in the SBR and A-trap was assumed to take place only through the excess sludge removal.

$E_{sludge\ transport}$ and $E_{urine\ transport}$ were the energy requirements for transporting of excess sludge from the UASB reactor and the SBR, and urine, respectively, from the on-site collection to agricultural land, and was calculated based on the primary energy requirement of 4.8 MJ/ton/km (including empty return trip) [217].

$E_{methane}$ was the energy produced as methane in the UASB reactor. The volume of produced methane was calculated from the COD load of the reactor, the methanization level of the influent and the theoretical methane production of 0.35 L/gCOD$_{converted}$. The methanization level of the influent was calculated as a mass proportion of the methanization levels of the sub-streams (70% for black water with kitchen refuse, 78% for feces with kitchen refuse [91] and 88% for grey water sludge [74]). The primary energy production from methane was calculated using the volume of methane and the calorific value of methane (35.8 MJ/m^3) [181].

The sludge production in the UASB reactor was calculated according to Zeeman and Lettinga (1999) [226] (Eq. 2.7)

$$X_p = O * SS * R * (1 - H) \tag{2.7}$$

where X_p is the sludge production (kgCOD/m^3/d), O is the organic loading rate (2.98 kgCOD/m^3/d [94]), SS is the fraction of suspended solids in the influent (COD$_{ss}$/ COD$_{total}$) (0.76 with a mixture of black water and kitchen refuse, and 0.88 with a mixture of feces and kitchen refuse [91]), R is the fraction of COD$_{ss\ removed}$ (0.96 [94]) and H is the level of hydrolysis of the removed solids (0.7 [91]). The total wet sludge production was calculated using the volume of the UASB reactor (calculations for the reactor volume is presented in the sub chapter *Calculations for reactor dimensions and land area requirement*) and the sludge concentration (34 gCOD/L [42]). The sludge production in the SBR was calculated using a sludge yield of 0.12 kgVSS/ kgCOD$_{removed}$ and a sludge concentration of 5.5 gVSS/L [75]. The sludge production in the A-trap was calculated using a sludge yield of 0.73 kgVSS/kgCOD$_{removed}$ and a sludge concentration of 6.3 gVSS/L [45]. The sludge production in the MBR was calculated from the flow mass balance of the system using a SRT of 1 d and HRT of 1.9 h [74].

2.2.6 Calculations for chemical use

In concepts 1 and 2 polymers were used for sludge dewatering and calcium oxide (CaO) was used for flu gas treatment after sludge incineration. The dose of CaO was 30 kg/tonDM and the dose of polymers was 7.1 kg/tonDM [78]. Methanol (CH_3OH) was consumed 1.48 kg/cap/year in the post-denitrification step in concept 2 [217]. In concepts 3 and 4 sodium hydroxide (NaOH) and magnesium chloride ($MgCl_2$) were used in struvite precipitation to increase the pH and the supersaturation state. Consumption of NaOH was calculated using Eq. 2.8

$$m_{NaOH} = M_{NaOH} * 10^{-14}(10^{pH_b} - 10^{pH_a}) \tag{2.8}$$

where m_{NaOH} is the mass of NaOH (g/L), M is the molecular mass (g/mol), pH_a is the influent pH of 7.7 [94] and pH_b is the operational pH of 9 [43]. Consumption of 33% NaOH was further determined from the mass of NaOH. Consumption of $MgCl_2$ was calculated from the influent phosphate concentration using a Mg/PO_4–P ratio of 1.5 [43].

2.2.7 Calculations for reactor dimensions and land area requirement

Total land area requirement for concepts 1 and 2 consisted of the volume of the biological reactors, secondary settling tank, digester, biogas storage tank and the urine storage tank (concept 2). The land use of the incineration process was not taken into account due to lack of data. Total land area requirement for concepts 3 and 4 consisted of the volume of the buffer tank (for UASB, SBR, A-trap and MBR), reactors (UASB, OLAND, Struvite, black water TF, SBR/A-trap/MBR and grey water TF), biogas storage tank and the urine storage tank (concept 4).

The volume of the biological reactors and secondary settling tank were according to Wilsenach and van Loosdrecht (2006) [217] and the volume of the buffer tanks, urine storage tank and reactors (Digester/UASB, Struvite, MBR and A-trap) were determined using the influent flow rate and the storage time or the HRT. The volume of the A-trap consisted of three parts: aerated grit chamber, A-trap reactor and settling tank. The storage time was 1 d for the UASB buffer tank (assumed), 0.3 d for the SBR, A-trap and MBR buffer tanks (assumed) and 6 months for the urine collection tank [122]. The HRT was 15 d for the digester [217], 0.08 d for the struvite reactor [43], 1.9 h for the MBR [74], 4 min and 54 min for the aerated grit chamber and settling tank, respectively [49], and 1.9 h for the A-trap reactor [45]. The HRT of the UASB reactor was calculated according to Zeeman and Lettinga (1999) [226] (Eq. 2.9)

$$HRT = C * \frac{SS}{X} * R * (1 - H) * SRT \tag{2.9}$$

where C is the influent COD_{total} concentration (gCOD/L), X is the sludge concentration in the reactor (34 gCOD/L [42]), SS is the fraction of suspended solids in the influent ($\mathrm{COD}_{ss}/\mathrm{COD}_{total}$) (0.76 with a mixture of black water and kitchen refuse, and 0.88 with a mixture of feces and kitchen refuse [91]), R is the fraction of $\mathrm{COD}_{ss\ removed}$ (0.96 [94]), H is the level of hydrolysis of the removed solids (0.7 [91]), and SRT is the sludge retention time (d) calculated from the sludge production (kgCOD/m^3/d) and the sludge concentration in the reactor.

The volume of the biogas storage tank was calculated using the volume of produced methane, the fraction of methane in biogas (65% [181]) and storage time of 1 d [91]. The volume of the SBR was calculated using the volume of wastewater per cycle (360 min) and a $\mathrm{volume}_{wastewater}/\mathrm{volume}_{total}$ ratio of 0.3 m^3/m^3 [181]. The volume of a single-stage TF was determined according to Tchobanoglous et al. (2004) [181] (Eq. 2.10),

$$V = \frac{W}{(\frac{100}{e*(1+0.4432)})^2} \tag{2.10}$$

where W is the BOD_5 loading and e is the BOD_5 removal efficiency. The depth of the filter was set to 2.1 m as the average depth in standard rate filters.

The volume of the OLAND reactor was determined from the length, width and height of the reactor. The length of the reactor was determined by the length of the shaft and the width and height by the disc diameter. To calculate the length of the shaft, the total number of discs was defined from the total surface area of discs and the disc diameter (determined previously with the energy requirement of OLAND). The length of the shaft was calculated using a disc thickness of 0.5 cm and a disc interspace of 1 cm [212]. The length, width and height of the reactor was then determined using the length of the shaft and the disc diameter, respectively, with 15% of the disc diameter as extra space.

Height of the buffer tanks, digester, UASB reactor and SBR was calculated using

Eq. 2.11 that was derived from the equation for cylinder volume using f as a height/diameter ratio.

$$H = \sqrt[3]{\frac{4 * V_{cylinder} * f^2}{\pi}}, \; f = \frac{H}{d} \tag{2.11}$$

where $V_{cylinder}$ is the volume of the reactor and f is the height/diameter ratio that was assumed to be 3 with the exception of the SBR with ratio of 1. The diameter was calculated using an assumed maximum height of 5 m as a boundary condition.

Height of the aerated grit chamber and settling tank of A-trap was calculated using Eq. 2.12 that was derived from the sum of cube volume and pyramid volume using f as $\text{height}_{pyramid}/\text{height}_{vessel}$ ratio.

$$H = \frac{V_{vessel}}{A * (1 - \frac{2}{3} * f)}, \; f = \frac{H_{pyramid}}{H_{vessel}} \tag{2.12}$$

V_{vessel} is the volume of aerated grit chamber and settling tank, A is the surface area and f is the $\text{height}_{pyramid}/\text{height}_{vessel}$ ratio of 0.1 for the aerated grit chamber and 0.5 for the settling tank. The surface area of the aerated grit chamber was calculated using a maximum surface loading of 30 m^3/m^2h and the surface area of the settling tank was calculated using a maximum surface loading of 1.5 m^3/m^2h [49]. Height of the A-trap reactor was considered to be the difference between height of the vessel and height of the pyramid.

2.3 Results and Discussion

2.3.1 Energy balance

Figure 2.2 presents the total primary energy consumption in the sanitation concepts. The highest primary energy consumption of 914 MJ/cap/year is attained in the centralized sanitation concept (concept 1), and by applying urine separation within the centralized concept, the primary energy consumption is decreased to 687 MJ/cap/year, creating a yearly energy saving of 227 MJ/cap. The lowest primary energy consumption of 437 MJ/cap/year is attained in the source-separation of urine, feces, kitchen refuse and grey water (concept 4 vacuum) using the A-trap for grey water treatment. Urine separation in the source-separation based sanitation concept creates a yearly energy saving of 200 MJ/cap using the SBR, 180 MJ/cap using the A-trap

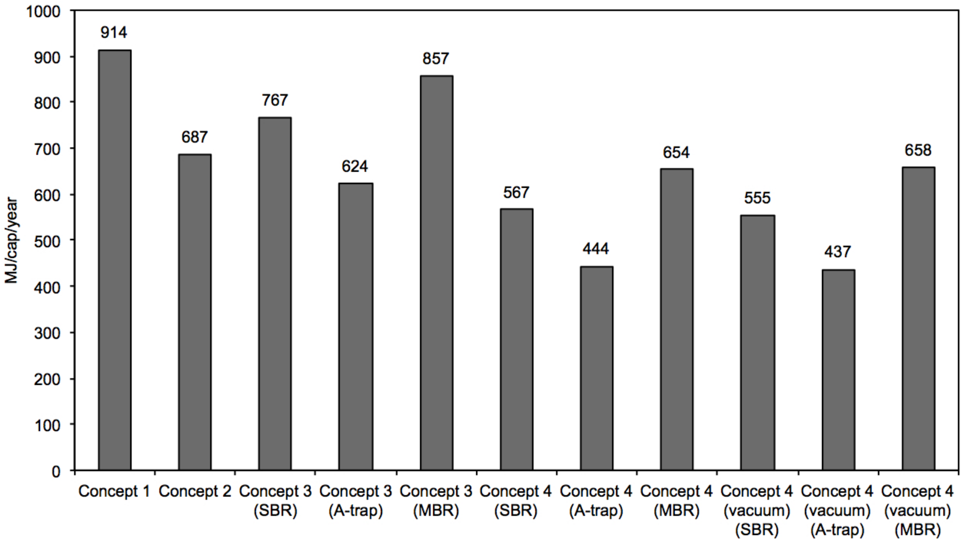

Figure 2.2: *Total primary energy consumption in sanitation concepts with different grey water treatment configurations*

and 203 MJ/cap using the MBR in concept 4 with gravity separation, and 212 MJ/cap using the SBR, 187 MJ/cap using the A-trap and 200 MJ/cap using the MBR in concept 4 with vacuum separation. Bioflocculation of grey water in the A-trap and subsequent grey water sludge co-digestion in the UASB reactor creates a yearly energy saving of 143 MJ/cap in concept 3, 123 MJ/cap in concept 4 (gravity) and 118 MJ/cap in concept 4 (vacuum) compared to the use of the SBR for grey water treatment. The high primary energy consumption of concept 1 originates mainly from the high energy input to mineralize organic matter in the AS process and the resulting low energy recovery as methane. The low primary energy consumption of concept 4 originates from the low water consumption of the urine diverting toilets, resulting in low energy demand of collection and treatment of feces and kitchen refuse. In addition, by grey water sludge co-digestion in the UASB reactor, high energy consumption for sludge transport can be avoided while simultaneously increasing energy recovery as methane.

The energy parameters together with the sludge production and urine collection for concepts 1 and 2 are presented in Table 2.4. The most prominent parameters in the energy balance in concepts 1 and 2 are energy consumption for the collection of wastewater and aeration of the biological reactors. The collection contributes 27% in concept 1 and 30% in concept 2 to the total primary energy consumption, and the aeration contributes 40% in concept 1 and 23% in concept 2. Furthermore, transporting of urine in concept 2 contributes 18% to the total primary energy consumption. Due to the shorter SRT in the A-trap (0.8 d) compared to the AS process (12 d), the energy consumption for aeration is significantly lower in concept 2 compared to concept 1. However, short SRT increases the excess sludge production leading to an increase in the energy requirement for heating of the digester. Nevertheless, the higher excess sludge production together with the low mineralization of organic matter creates almost twice as high methane production in concept 2 compared to concept 1. Compared to the study of Wilsenach and van Loosdrecht (2006) [217], both concepts have higher total primary energy consumption mainly due to the energy consumption for the collection that is included in this study and the higher energy consumption for the transporting of collected urine compared to the treatment of urine and sludge rejection water in struvite precipitation and SHARON processes, used in the study of Wilsenach and van Loosdrecht (2006) [217]. However, direct reuse of urine provides a clean route for nutrient recovery, while the mixing of sludge rejection water with urine might deteriorate the quality of the produced struvite with heavy metals from sewage.

Table 2.4: *Sludge production, urine collection and energy consumption and production (methane) in concepts 1 and 2 (primary energy presented as bolded figures)*

Parameter	Unit	Concept 1	Concept 2
Urine collection	kg/cap/y	-	743
Sludge production	kgWS/cap/y	1048	1201
	kgDS/cap/y	131	150
$E_{collection}$	kWh/cap/y	25	25
	MJ/cap/y	**288**	**288**
$E_{aeration}$	MJ/cap/y	135	68
	MJ/cap/y	**432**	**218**
E_{mixing}	MJ/cap/y	37	17
	MJ/cap/y	**118**	**54**
$E_{pumping}$	MJ/cap/y	20	15
	MJ/cap/y	**64**	**48**
$E_{heating(digester)}$	**MJ/cap/y**	**104**	**114**
$E_{dewatering}$	**MJ/cap/y**	**5**	**5**
$E_{sludge\ transport}$	**MJ/cap/y**	**6**	**7**
$E_{incineration}$	**MJ/cap/y**	**54**	**52**
$E_{urine\ transport}$	**MJ/cap/y**	**-**	**178**
$E_{methane}$	**MJ/cap/y**	**157**	**277**
E_{total}	**MJ/cap/y**	**914**	**687**

WS=Wet Sludge
DS=Dry Sludge

The energy parameters together with the sludge production, urine collection and UASB influent characteristics for concepts 3 and 4 are presented in Table 2.5 and 2.6. The most prominent parameters in the energy balance in concepts 3 and 4 are energy consumption for the vacuum collection and transport of black water and kitchen refuse, and heating of the UASB reactor. The vacuum collection and transport contributes 27–35% in concept 3 and 15–20% in concept 4 (vacuum) to the total primary energy consumption, and heating of the UASB reactor contributes 33–46% in concept 3, 36–53% in concept 4 (gravity) and 24–43% in concept 4 (vacuum). Furthermore, transporting of collected urine in concept 4 contributes 17–23% to the total primary energy consumption.

Table 2.5: *UASB influent characteristics, sludge production and urine collection in concepts 3 and 4 with different grey water treatment configurations (without co-digestion using the SBR or with co-digestion using the A-trap/MBR) (UASB=Upflow Anaerobic Sludge Blanket reactor, OLAND=Oxygen Limited Anaerobic Nitrification Denitrification, Struvite precipitator, TF=Trickling Filter, SBR=Sequencing Batch Reactor (SBR), A-trap=A-stage of AB-process and MBR=Membrane Bio Reactor)*

Parameter	Unit	Concept 3			Concept 4					
					Gravity toilet			Vacuum toilet		
		SBR	A-trap	MBR	SBR	A-trap	MBR	SBR	A-trap	MBR
UASB INFLUENT										
Volume	m^3/cap/y	3	4	5	2	3	4	1	2	3
Temperature	°C	16	15	13	12	11	11	11	11	10
Methanization level	%	70	79	80	78	79	80	78	79	80
SLUDGE PRODUCTION										
UASB reactor	kg/cap/y	277	321	365	299	343	394	299	343	394
SBR/A-trap/MBR	kg/cap/y	373	682	2128	373	682	2128	373	682	2128
URINE COLLECTION	kg/cap/y	-	-	-	743	743	743	743	743	743

Table 2.6: *Energy consumption and production (methane) in concepts 3 and 4 (primary energy presented as bolded figures) (UASB=Upflow Anaerobic Sludge Blanket reactor, OLAND=Oxygen Limited Anaerobic Nitrification Denitrification, Struvite precipitator, TF=Trickling Filter, SBR=Sequencing Batch Reactor (SBR), A-trap=A-stage of AB-process and MBR=Membrane Bio Reactor)*

Parameter	Unit	Concept 3			Concept 4					
					Gravity toilet			Vacuum toilet		
		SBR	A-trap	MBR	SBR	A-trap	MBR	SBR	A-trap	MBR
$E_{collection}$	kWh/cap/y	30	30	30	5	5	5	13	13	13
	MJ/cap/y	**346**	**346**	**346**	**58**	**58**	**58**	**150**	**150**	**150**
$E_{heating(UASB)}$	**MJ/cap/y**	**341**	**422**	**584**	**305**	**385**	**547**	**199**	**280**	**441**
E_{OLAND}	kWh/cap/y	1.3	1.6	2.2	0.2	0.3	0.4	0.3	0.4	0.8
	MJ/cap/y	**15**	**18**	**25**	**2**	**3**	**5**	**3**	**5**	**9**
$E_{Struvite}$	kWh/cap/y	4.4	5.4	7.5	0.8	1.0	1.5	0.9	1.5	2.7
	MJ/cap/y	**51**	**62**	**86**	**9**	**12**	**17**	**10**	**17**	**31**
$E_{TF\ (BW)}$	kWh/cap/y	0.2	0.3	0.4	0.2	0.2	0.3	0.1	0.1	0.2
	MJ/cap/y	**2**	**3**	**5**	**2**	**2**	**3**	**1**	**1**	**2**
$E_{sludge\ transport}$	**MJ/cap/y**	**156**	**77**	**88**	**161**	**83**	**95**	**161**	**83**	**95**
E_{SBR}	MJ/cap/y	33	-	-	33	-	-	33	-	-
	MJ/cap/y	**106**	**-**	**-**	**106**	**-**	**-**	**106**	**-**	**-**
E_{A-trap}	MJ/cap/y	-	7.2	-	-	7.2	-	-	7.2	-
	MJ/cap/y	**-**	**23**	**-**	**-**	**23**	**-**	**-**	**23**	**-**
E_{MBR}	kWh/cap/y	-	-	8.7	-	-	8.7	-	-	8.7
	MJ/cap/y	**-**	**-**	**100**	**-**	**-**	**100**	**-**	**-**	**100**
$E_{TF\ (GW)}$	kWh/cap/y	2.1	2.1	2.1	2.1	2.1	2.1	2.1	2.1	2.1
	MJ/cap/y	**24**	**24**	**24**	**24**	**24**	**24**	**24**	**24**	**24**
$E_{urine\ transport}$	**MJ/cap/y**	**-**	**-**	**-**	**178**	**178**	**178**	**178**	**178**	**178**
$E_{methane}$	**MJ/cap/y**	**274**	**352**	**401**	**278**	**324**	**373**	**278**	**324**	**373**
E_{total}	**MJ/cap/y**	**767**	**624**	**857**	**567**	**444**	**654**	**555**	**437**	**658**

Urine separation in the source-separation based sanitation concept (concept 4) has the potential to decrease the total energy consumption due to a lower energy demand of the feces collection and the post-treatment of UASB reactor effluent in the OLAND reactor, struvite precipitator and TF compared to concept 3. In addition, separation of urine from feces and kitchen refuse and the low water consumption of the urine diverting toilets decreases the UASB reactor influent volume and thus the energy used for heating of the reactor. However, urine separation has an extra energy consumption for transporting of collected urine. Although vacuum collection of feces and kitchen refuse increases the energy demand of collection compared to gravity collection, vacuum separation of urine presents the energetically most favorable option due to the smallest UASB reactor influent volume.

Significant fraction of the energy consumption for the SBR originates from the high aeration demand at the long SRT (15 d [75]). By decreasing the SRT to 0.6 d using the A-trap [45] or to 1 d using the MBR [74]), the energy consumption for the grey water treatment system can be decreased. The energy consumption for the MBR, however, is four times higher than for the A-trap due to the higher energy requirement of membrane technology. When grey water sludge is co-digested in the UASB reactor, the total energy consumption can be decreased as no transporting of grey water sludge is required. Furthermore, methane production in the UASB reactor can be increased due to the higher loading of the reactor and the higher methanization level of grey water sludge compared to black water, feces and kitchen refuse. However, co-digestion of grey water sludge increases the heating energy required for the reactor as a result of a higher influent volume and a lower influent temperature, originating from the lower grey water sludge temperature that was assumed to be the environmental temperature. Consequently, bioflocculation of grey water in the MBR and subsequent grey water sludge co-digestion in the UASB reactor is not energetically favorable compared to grey water treatment in the SBR, due to the high sludge production in the MBR, and the resulting high heating energy requirement for the UASB reactor. However, to decrease the volume of the MBR sludge, a settler can be implemented to increase the concentration of the sludge.

2.3.2 Water reuse

Table 2.7 presents the calculated effluent quality of the different grey water treatment systems and the standards for non-potable grey water reuse suggested by Li et al. (2009) [108]. The reuse standards were divided into recreational impoundments, such as ornamental fountains and lakes, and urban reuse, such as toilet flushing, laundry and irrigation. Unrestricted reuse is considered in close contact with people and restricted reuse in areas without public access. Due to high nutrient concentrations in the effluent, none of the treatment systems fulfilled the reuse standards for recreational impoundments. The SBR and the MBR with TF as a post-treatment step fulfilled the standards for urban reuse, but only the effluent from the SBR-TF was according to the unrestricted reuse. The better effluent quality from the SBR-TF in terms of BOD_5 can be explained by the longer SRT and thus, more extensive degradation of organic material. However, membrane technology has the potential to produce grey water effluent free of solids and therefore benefit the use of advanced post-treatment systems such as UV and ozonation for removing micro-pollutants and pathogens. Nevertheless, the costs of advanced post-treatment systems have to be related to the actual need for high quality water rather than striving to fulfill the most stringent standards.

Table 2.7: *Calculated effluent quality of grey water treatment systems and suggested standards for water reuse (Unit mg/L)*

Parameter	Grey water effluent quality (This study)			Suggested reuse standards [108]			
				Recreational impoundments		Urban reuse	
	SBR -TF	A-Trap -TF	MBR -TF	Restrict.	Unrestrict.	Restrict.	Unrestrict.
BOD_5	5	30	14	30	10	30	10
TSS	25	60	6	30	-	30	-
TN	10	10	3	1	1	-	-
TP	4	3	2	0.05	0.05	-	-

2.3.3 Nutrient recovery

Nutrients such as nitrogen, phosphorus and potassium can be recovered using urine separation in the centralized concept and in the source-separation based sanitation concepts. Nutrients were considered to be recovered through urine spreading on agricultural land in concepts 2 and 4, and thus, all the nutrients present in the collected urine (collection degree of 75%) were considered to be recovered. Struvite ($MgNH_4PO_4*6H_2O$) precipitation is used to recover nutrients from the effluent of the OLAND reactor in concepts 3 and 4. Struvite is produced 2.13 kg/cap/year from which 0.27 kg is phosphorus and 0.12 kg is nitrogen in concept 3, and 1.0 kg/cap/year from which 0.13 kg is phosphorus and 0.06 kg is nitrogen in concept 4. In concepts 3 and 4 nutrients were also considered to be recovered from the excess sludge of the UASB reactor and the SBR through sludge reuse on agricultural land. Nitrogen and phosphorus removed in the UASB reactor and the SBR were considered to be trapped in the sludge and this way to be recovered. Figure 2.3 presents the nutrients recovered in concepts 2, 3 and 4 with different grey water treatment configurations. As most of the nutrients are present in urine, source-separation and direct reuse of urine brings forth a major contribution to the total nutrient recovery. The choice between the different grey water treatment configurations (SBR/A-trap/MBR) has only a slight effect on the total amount of nutrients recovered. The maximum nutrient recovery can be achieved with concept 4, where nutrient recovery from sludge increases the recovery of nitrogen and phosphorus compared to concept 2.

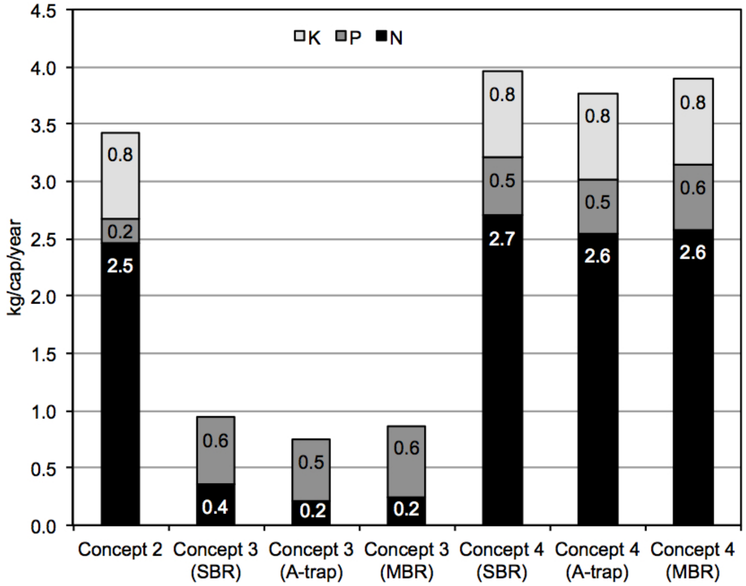

Figure 2.3: *Nutrient recovery in concepts 2, 3 and 4 with different grey water treatment configurations*

Compared to artificial fertilizers, direct reuse of urine in agriculture, as suggested here, has an advantage of acting as a multicomponent fertilizer. However, direct reuse of urine also has disadvantages, such as transporting of urine to agricultural land and the possible adverse effect of high salt content of urine on soil, especially in low rainfall areas. Several technologies have been presented to overcome these issues by indirectly recovering the resources from urine. Nutrients can be recovered from urine by struvite precipitation [101][122] or using algae for nutrient up-take from urine and subsequent reuse of algae biomass [2]. In the study of Kuntke et al. (2012) [96] a microbial fuel cell was used to simultaneously produce energy (3.46 kJ/gN) and recover ammonium (3.29 gN/d/m^2) from urine. By replacing the urine transport with a microbial fuel cell, the total primary energy consumption can be decreased by 19% in concept 2 and 17–23% in concept 4, indicating a promising new direction for urine treatment.

According to the current Dutch guidelines for sewage sludge reuse in agriculture (BOOM), reuse of black water sludge is prohibited due to elevated concentrations of copper and zinc [41]. However, as black water is predominantly human originated (urine, feces and tap water), the applicability of sewage sludge reuse guidelines on the reuse of black water sludge can be argued. Furthermore, the amount of heavy metals related to the phosphorus content of sludge is significantly higher in cow manure [202] and in artificial phosphorus fertilizers in the case of cadmium, chromium and nickel [149]. The heavy metal content of grey water sludge and the effect of grey water sludge co-digestion on the excess sludge quality of UASB reactor needs to be further investigated to decide whether or not to mix these streams.

2.3.4 Energy balance including water saving and reuse and nutrient recovery

Compared to the normal flush toilet in concept 1, the use of a urine diverting toilet or a vacuum toilet saves water of a drinking quality. The vacuum toilet saves 28 L/cap/day, the urine diverting toilet (gravity) saves 29 L/cap/day and the urine diverting toilet (vacuum) saves 32 L/cap/day. Considering a primary energy consumption of 5.4 MJ/m^3 for drinking water production and distribution [58] (using efficiency of 0.31 [190]), 57 MJ/cap/year can be indirectly gained in concepts 2 and 4 (gravity), 55 MJ/cap/year in concept 3 and 63 MJ/cap/year in concept 4 (vacuum). Furthermore, by reusing grey water effluent for toilet flushing, laundry and irrigation, drinking water can be saved and energy can be indirectly gained in concepts 3 and 4. By assuming full reuse of grey water effluent (29 m^3/cap/year), energy can be indirectly gained 157 MJ/cap/year by using either the SBR-TF for unrestricted or the MBR-TF for restricted urban reuse. As the water use for toilet flushing and laundry is only 8 m^3/cap/year [27], 73% of the SBR-TF effluent is left for irrigation. Grey water effluent from the MBR-TF can only be used for urban reuse applications without public access, such as irrigation of restricted areas.

Through the recovery of nutrients, energy can be indirectly gained in the production of artificial fertilizers. Considering a primary energy requirement of 45 MJ/kgN, 29 MJ/kgP and 11 MJ/kgK for fertilizer production [122], energy can be indirectly gained 129 MJ/cap/year in concept (2), in concept (3) 33 MJ/cap/year with SBR, 25 MJ/cap/year with A-trap, 29 MJ/cap/year with MBR, in concept (4) 145 MJ/cap/year with SBR, 137 MJ/cap/year with A-trap and 141 MJ/cap/year with MBR.

Figure 2.4 presents the total primary energy consumption with and without the indirect energy gain from water saving and reuse, and nutrient recovery. The most prominent energy gain can be achieved with the recovery of nutrients through urine separation (concepts 2 and 4) and the reuse of grey water effluent using either the SBR or the MBR (concepts 3 and 4). Due to the significant energy gain from the grey water effluent reuse, grey water treatment in the SBR becomes energetically more favorable than bioflocculation of grey water in the A-trap and subsequent grey water sludge co-digestion in the UASB reactor. Beside water and nutrient recovery, there is an increasing interest to recover the heat content of wastewater [205]. Heat recovery on-site from source-separated grey water using a heat exchanger would be an energy efficient option to preheat the incoming tap water as no electricity is needed.

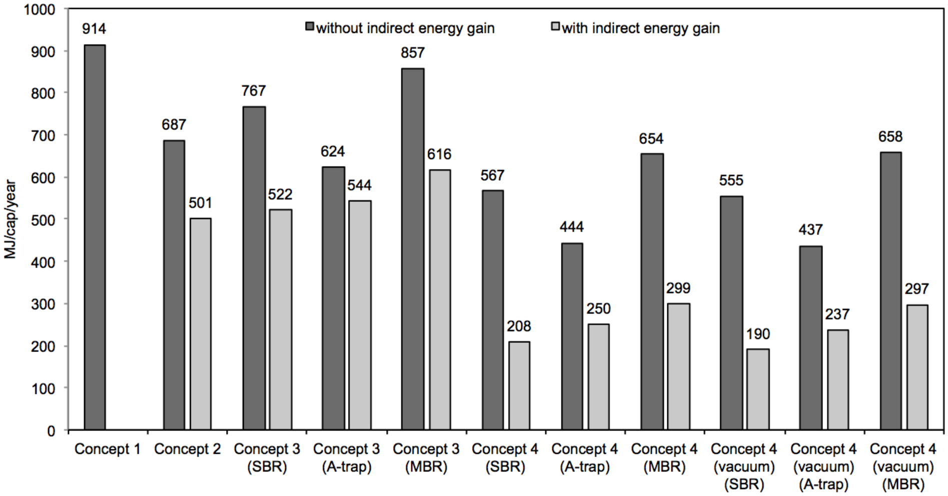

Figure 2.4: *Total primary energy consumption in sanitation concepts with and without indirect energy gain from water saving and reuse, and nutrient recovery*

When the indirect energy gain is taken into account, urine separation applied in the centralized sanitation creates even higher yearly energy saving of 413 MJ/cap compared to concept 1. The lowest energy consumption in concept 3 (522 MJ/cap/year) and concept 4 (208 MJ/cap/year (gravity) and 190 MJ/cap/year (vacuum)) is attained when the SBR is used. By applying urine separation in the source-separation based sanitation, 294–331 MJ/cap/year can be saved with indirect energy gain.

2.3.5 Chemical use

Figure 2.5 presents the chemical use in concepts 1, 2, 3 and 4 with different grey water treatment configurations. The chemical use in concepts 1 and 2 is considerably higher than in concepts 3 and 4 due to the high sludge production in aerobic processes and the resulting consumption of polymers for sludge dewatering and CaO for flu gas treatment after sludge incineration. As the sludge production in concept 2 is higher than in concept 1 (due to the shorter SRT in the aerobic process), the chemical use is accordingly higher. Furthermore, additional chemical use in concept 2 originates from the consumption of methanol in the post-denitrification step. As the amount of NaOH is calculated to be negligible, the only chemical taken into account in the struvite precipitation in concepts 3 and 4 is $MgCl_2$. The use of $MgCl_2$ is the highest in concept 3, due to the highest phosphate concentration in the OLAND reactor effluent. Grey water treatment in the MBR and the subsequent grey water sludge co-digestion in the UASB reactor slightly increases the $MgCl_2$ consumption due to the increased phosphate loading. The use of either gravity or vacuum urine diverting toilet does not influence the chemical use in concept 4. Contrary to the centralized concept, urine separation in the source-separation based concept decreases the chemical use.

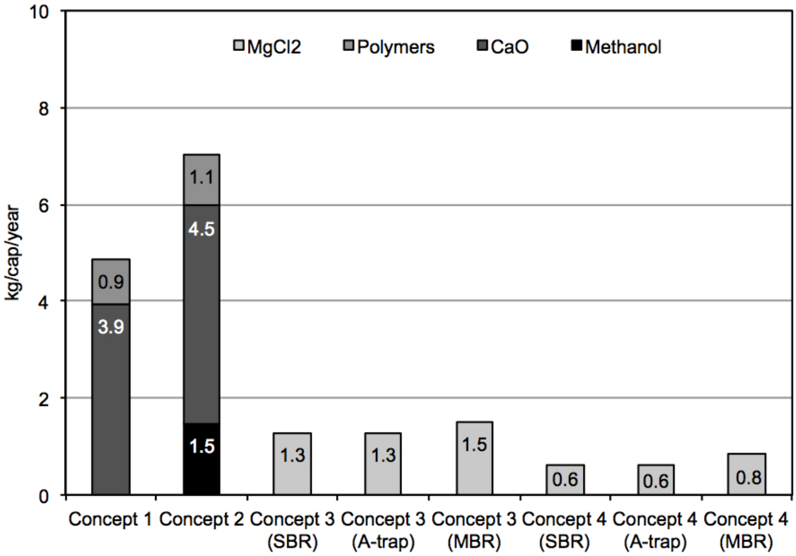

Figure 2.5: *Chemical use in concepts 1, 2, 3 and 4 with different grey water treatment configurations*

2.3.6 Effluent quality

Within the European Union the discharge of wastewater effluent is controlled by the pollutant removal efficiencies of the treatment systems and the final effluent concentrations per connected person according to the EU Water Framework Directive 91/ 271/EEC [56]. Table 2.8 presents the calculated effluent quality of the different sanitation concepts and the discharge standards. In concepts 3 and 4 only the effluent discharge of the source-separated concentrated stream is taken into account, leaving out the grey water effluent that is considered to be reused. For simplicity, the effluent quality presented in concepts 3 and 4 is the average of the different grey water treatment configurations (without co-digestion using the SBR or with co-digestion using the A-trap/MBR). The pollutant concentrations in the effluent of the concentrated stream are higher and the pollutant loadings are lower without grey water sludge co-digestion, due to the lower UASB reactor influent volume compared to co-digestion.

Table 2.8: *Calculated effluent quality in sanitation concepts and discharge standards*

Parameter	Unit	C1	C2	C3	C4	Discharge standards [56]
COD	mg/L	46	44	155	187	125
BOD_5	mg/L	6	24	83	100	25
TSS	mg/L	34	47	393	385	35
TN	mg/L	9	6	350	70	15
TP	mg/L	4	1	27	17	2
COD	g/cap/y	5037	4802	599	551	-
BOD_5	g/cap/y	657	2619	321	297	-
TSS	g/cap/y	3723	5129	1520	1148	-
TN	g/cap/y	986	655	1392	221	-
TP	g/cap/y	438	109	104	51	-

As the total pollutant removal efficiencies in concepts 3 and 4 are mostly higher than in concepts 1 and 2 (Table 2.3), the higher pollutant concentrations in the effluent in concepts 3 and 4 originate from the higher concentrations in the source-separated streams. Consequently, according to the current discharge standards that are based on pollutant concentrations rather than pollutant loadings, the discharge of effluent in concepts 3 and 4 is prohibited. However, as the pollutant loadings in the effluent in concepts 3 and 4 decrease by up to 90% compared to concepts 1 and 2, the future discharge standards ought to consider also the total pollutant load discharged from wastewater treatment. With urine separation (concepts 2 and 4) both nutrient (N and P) concentrations and loadings are decreased.

The COD/BOD ratio of the effluent loading in concept 1 is higher than in other concepts, originating from the high BOD_5 removal efficiencies in the existing wastewater treatment plants in the Netherlands, applied in concept 1. More data on the actual BOD_5 removal efficiencies in the A-trap in concept 2 and in the OLAND reactor in concepts 3 and 4 is required to confirm the actual COD/BOD ratio of the effluent loading. To deal with the current discharge standards, further treatment of effluent in concepts 3 and 4 need to be considered. However, according to the COD:N:P ratio of 100:20:1 necessary for biological treatment [181], the effluent is short in organic matter with a ratio of 100:226:18 (concept 3) and 100:38:9 (concept 4), and requires an alternative treatment method or a source of organic matter.

2.3.7 Land area requirement

The total volume of the treatment systems in concept 1 is 0.32 m^3/cap and in concept 2 is 0.53 m^3/cap, of which 0.38 m^3/cap originates from the urine storage tank. The total volume of the treatment systems for black water and kitchen refuse (concept 3) is 0.15–0.22 m^3/cap, and for feces and kitchen refuse (concept 4) is 0.13–0.17 m^3/cap, the lowest value being without grey water sludge co-digestion and the highest with grey water sludge co-digestion using the MBR for bioflocculation of grey water. Grey water treatment in the SBR-TF requires a total volume of 0.16 m^3/cap, the A-trap-TF requires 0.29 m^3/cap, and the MBR-TF requires 0.14 m^3/cap. The total volume of the treatment systems in concept 3 is 0.31–0.47 m^3/cap and in concept 4 is 0.67–0.81 m^3/cap, reaching highest volumes with the A-trap and lowest with the SBR. Urine separation in both centralized and source-separation based sanitation concepts increases the land area requirement due to the large volume of the urine storage tank. In addition, the land use of the incineration process (concept 1 and 2) will further increase the land area requirement. The lowest land area requirement is achieved with source-separation of black water and kitchen refuse and by using the SBR for grey water treatment.

2.3.8 Sensitivity analysis

The SRT applied in the high loaded biological reactors, such as the A-trap, can have significant influence on the pollutant removal efficiencies and resulting effluent quality. For example, the removal efficiencies of the A-trap used for sewage treatment in concept 2 are significantly higher than of the A-trap used for grey water treatment in concepts 3 and 4 (Table 2.3). The A-trap used for sewage treatment is according to the study of Wilsenach and van Loosdrecht (2006) [217] in which an SRT of 0.8 d was assumed to attain the highest effluent quality, while the SRT of the A-trap used for grey water treatment is according to the actual SRT of 0.6 d applied at the demonstration site of DeSaH B.V. (2010) [45], resulting in lower removal efficiencies similar to the ones reported by Böhnke (1981) [16]. Consequently, if the SRT of the A-trap for grey water treatment is increased to 0.8 d, the pollutant removal efficiencies could be increased, resulting in higher effluent quality. Furthermore, effluent from the A-trap with higher quality could be reused according to the urban reuse standards, resulting in a significant indirect energy gain from water reuse, and turning the use of the A-trap and subsequent grey water sludge co-digestion into an energetically more favorable option than the use of the SBR. However, due to limited experimental data and the different composition of grey water and sewage, more research is required to confirm the relation between the SRT of the A-trap and the pollutant removal efficiencies.

Significant part of the total energy consumption in the sanitation concepts originates from the energy used for heating the digester and the UASB reactor. Location specific data on the environmental temperature and the tap water temperature have a major effect on the energy demand of heating, as the tap water temperature defines the amount of energy used for heating up the influent and the environmental temperature defines the amount of energy used to compensate heat loss through reactor walls. For example, if the tap water and environmental temperature is increased to 15°C (as an average annual temperature in the south of Europe), the primary energy consumption for heating decreases by 13–20% in all sanitation concepts. In contrast, if the tap water and environmental temperature is decreased to 6°C (as an average annual temperature in the north of Europe), the primary energy consumption for heating increases by 15–21% in all sanitation concepts. The location and the according temperatures may therefore affect the feasibility of grey water sludge co-digestion in the UASB reactor, especially when grey water is concentrated in the MBR with a high sludge production.

The transport distance of urine and excess sludge is another location specific parameter significantly influencing the energy balance of the sanitation concepts. Accessibility and the demand for fertilizers on agricultural land in the vicinity determines the transport distance of urine and excess sludge. In the case of centralized sanitation, the critical distance to agricultural land at which urine transport (concept 2) becomes unfavorable compared to concept 1 is 410 km, including the indirect energy gain from water saving and nutrient recovery. This distance covers transport of urine from the Netherlands to France and is higher than any actual distance to accessible agricultural land. However, to avoid high energy consumption of transporting, collected urine should be concentrated at long distances. When considering the use of a vapour compression distillation process with an average primary energy consumption of 337 MJ/m^3 [123], the critical distance at which evaporation of urine becomes more favorable than transporting of urine is 90 km. In the case of source-separation based sanitation, the critical distance to agricultural land at which urine and excess sludge transport (from the UASB reactor and the SBR) becomes unfavorable compared to concept 1 is 140 km in concept 3 and 150 km in concept 4, including the indirect energy gain from water saving and reuse, and nutrient recovery. Furthermore, by using the A-trap for bioflocculation of grey water and subsequent grey water sludge co-digestion in the UASB reactor, the critical distance is increased to 300 km in concept 3 and to 180 km in concept 4, covering the transport within the Netherlands. Although the transport of urine and excess sludge over long distances is never the optimal solution for nutrient recovery, the long critical distances presented above realizes the possibilities of implementing nutrient recovery technologies in locations surrounded by agricultural lands with surplus of nutrients.

According to the study of Thibodeau et al. (2011) [186], one of the most critical factors influencing the economic viability of source-separation of black and grey water is the water consumption for vacuum toilet. Reduction in the vacuum toilet flow has a major effect not only on the heating energy used for the UASB reactor, but also on the energy consumption for the vacuum collection and transport of wastewater. For example, if the water consumption for the vacuum toilet used for black water is decreased to 1.5 L/cap/d (0.25 L per flush) and the energy consumption for the vacuum collection is assumed to decrease by 75% ($\frac{1.5}{6}$ L), the energy consumption in concept 3 can be decreased by 35–55%, attaining the lowest primary energy consumption (156 MJ/cap/year using the SBR) of all the sanitation concepts, including the indirect energy gain from water saving and reuse, and nutrient recovery.

2.3.9 Outlook

This study provides insight into the influence of urine separation and different grey water treatment configurations (with (A-trap/MBR) and without (SBR) grey water sludge co-digestion) on the energy and material balances of centralized and source-separation based sanitation concepts. The energy and material balances are based on collection, transport and treatment of wastewater leaving out the energy and materials used in the construction and maintenance of the required infrastructure. However, according to Tidåker et al. (2007) [187], the energy use for the source-separation infrastructure is significant, and further research is therefore needed to complete the total life cycle of the sanitation concepts.

This study emphasizes the direct reuse of source-separated urine as a multicomponent fertilizer in agriculture. Beside the downside of urine transport, direct reuse also involves concerns about the contamination of soil and plants by pharmaceutical residues present in urine [221]. Further research on technologies for indirect resource recovery from urine would help to address both of these issues. Nevertheless, micro-pollutants are widely measured also from wastewater effluents and receiving water bodies, posing an actual contamination risk on the surrounding agriculture and drinking water production [182]. Clearly, micro-pollutants are of concern not only in the reuse of source-separated waste streams, but in the whole urban water cycle.

As most of the pathogens in wastewater are present in the black water stream, the pathogenic risk related to land application of black water sludge should be minimized. This could be done by pasteurizing black water in a hyper-thermophilic UASB reactor at 70°C [224]. Since the higher operational temperature of the UASB reactor will increase its energy demand, the water consumption for vacuum collection of black water should be minimized. Alternatively, black water sludge could be disinfected by co-composting with locally available garden waste.

To guarantee the optimal energy recovery from domestic wastewater streams, the influence of grey water sludge co-digestion on the UASB reactor performance, in particular the effect of surfactants on the digestion process needs to be further investigated. In addition, the effect of grey water sludge co-digestion on the excess sludge quality in terms of heavy metals and micro-pollutants should be determined.

Beside struvite recovery, further research should focus on alternative phosphorus recovery technologies to minimize the chemical use and to produce other phosphorus products, such as calcium phosphate, more suited for the needs of current fertilizer industries. Furthermore, to promote the full closing of carbon and nutrient cycles, a better understanding on the origin of heavy metals in the excess sludge of the UASB reactor is required. By targeted and functional standards for the sludge reuse in agriculture, resources from the source-separated waste streams can be recovered in such a way that the soil quality is improved.

2.4 Conclusions

The highest primary energy consumption of 914 MJ/cap/year is attained within the centralized sanitation concept. By coupling the centralized concept with source-separation of urine the energy consumption is decreased to 687 MJ/cap/year, and further to 501 MJ/cap/year with indirect energy gain from water saving and nutrient recovery.

Source-separation of black water, kitchen refuse and grey water results in a primary energy consumption of 767 MJ/cap/year, and in a consumption of 522 MJ/cap/year with indirect energy gain from water saving and reuse, and nutrient recovery. Urine separation within the source-separation based sanitation concept decreases the energy consumption to 567 MJ/cap/year with a gravity urine diverting toilet and to 555 MJ/cap/year with a vacuum urine diverting toilet. With the indirect energy gain from water saving and reuse, and nutrient recovery, the energy consumptions are further decreased, reaching the lowest energy consumptions of 208 MJ/cap/year (gravity) and 190 MJ/cap/year (vacuum) of all the sanitation concepts.

Source-separation of urine not only improve the energy balance and nutrient recovery, but also increases the effluent quality in terms of nutrient concentrations and the overall pollutant loading in both centralized and source-separation based sanitation concepts. However, larger land area and higher chemical use in the centralized concept is required.

Grey water bioflocculation in the A-trap and subsequent grey water sludge co-digestion in the UASB reactor decreases the primary energy consumption by 19% in the source-separation of black water, and 22% (gravity) and 21% (vacuum) in the source-separation of urine and feces, compared to grey water treatment in the SBR without grey water sludge co-digestion. However, as grey water effluent from the A-trap does not comply with the water reuse standards, in contrast to effluent from the SBR, the use of the SBR for grey water treatment becomes energetically more favorable than the A-trap when indirect energy gain from water reuse is taken into account. Although grey water effluent from the MBR is applicable for water reuse, the high sludge production and the resulting high energy consumption makes the use of the MBR energetically unfavorable.

Acknowledgements

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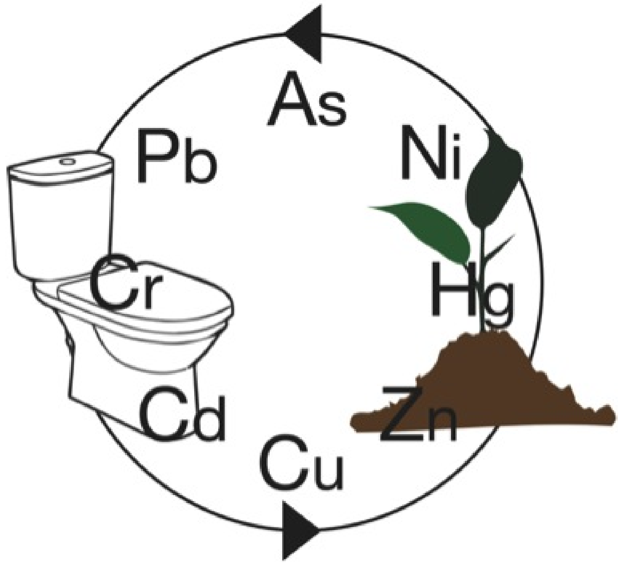
Chapter 3

Black water sludge reuse in agriculture: Are heavy metals a problem?



Abstract

Heavy metal content of sewage sludge is currently the most significant factor limiting its reuse in agriculture within the European Union. In the Netherlands most of the produced sewage sludge is incinerated, mineralizing the organic carbon into the atmosphere rather than returning it back to the soil. Source-separation of black water (toilet water) excludes external heavy metal inputs, such as industrial effluents and surface run-offs, producing sludge with reduced heavy metal content that is a more favorable source for resource recovery. The results presented in this paper show that feces is the main contributor to the heavy metal loading in vacuum collected black water (52%–84%), while in sewage the contribution of feces is less than 10%. To distinguish these two streams in the sludge reuse regulation, a control parameter should be implemented, such as the Hg and Pb content that is significantly higher in sewage sludge compared to black water sludge (from 50– to 200–fold). The heavy metals in feces and urine are primarily from dietary sources, and promotion of the soil application of black water sludge over livestock manure and artificial fertilizers could further reduce the heavy metal content in the soil/food cycle.

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3.1 Introduction

Soil is an important carbon storage and can hold three times the amount of carbon present in the atmospheric carbon pool [97]. Even a minute change in soil carbon reserve could therefore result in a significant change in the atmospheric CO_2 concentration [37]. The carbon sink capacity of soil impacts significantly not only the global climate change but also the world food security [98]. Soil erosion and microbial mineralization of organic carbon due to land use change and soil cultivation are suggested as the major routes for soil organic carbon loss [39]. To restore the soil organic carbon pool, reuse of crop residues and bio-solids, such as compost and manure, in agriculture is promoted [97].

Soil application of sewage sludge is considered as one of the most desired disposal methods to utilize its rich organic and inorganic plant nutrient content, but the presence of potentially toxic metals often restricts its use [168]. Reuse of sewage sludge in agriculture therefore divides opinions and the legislation regulating its use is highly diverse in different regions [150]. Within the European Union (EU) the soil application of sewage sludge is regulated under the Directive 86/278/EEC that sets the minimum quality standards for the soil and sludge used in agriculture in terms of heavy metal concentration and load [53]. However, several member states of the EU define national limit values apart from the EU standards, resulting in a great variety of sewage sludge treatment and disposal practices [87]. The Netherlands, being among the most rigid of the member states, sets the limit values below the common EU standards, prohibiting the soil application of sewage sludge [17]. Instead, the most common disposal route is incineration that not only mineralizes the organic carbon into CO_2 and destroy the plant nutrients nitrogen and phosphorus, but also requires additional energy input [58].

Source-separation of black water (toilet water) excludes external heavy metal inputs, such as industrial effluents and surface run-offs, and is therefore a more favorable source for resource recovery compared to sewage [220]. Vacuum collection of black water and sub-sequent treatment in an upflow anaerobic sludge blanket (UASB) reactor produces sludge with minimum input of heavy metals from household chemicals [225]. Although the heavy metal content is significantly lower compared to sewage sludge, the reuse of black water sludge is prohibited in the Netherlands under the Dutch guidelines due to elevated Cu and Zn concentrations [41]. However, as the characteristics of sewage and concentrated black water are different, leading to different origin of heavy metals in these streams, the use of the same guidelines on both streams can be argued.

There is a need to work towards a more targeted regulation of the soil application of wastewater sludge that is based on the characteristics and the origin of heavy metals in the sludge. Several studies have been conducted to determine the origin of heavy metals in municipal sewage [130, 31, 90, 170, 155] and source-separated domestic wastewater (black water/feces and urine, grey water and solid bio-waste [204, 143, 206]. No studies, however, have drawn the full heavy metal mass balance of black water including the fractions of toilet paper and toilet detergent, and investigate the primary origin of heavy metals in the black water components. Furthermore, no studies have compared the heavy metal content of black water and sewage sludge, and critically evaluated the applicability of current sludge reuse regulation on these different streams.

This study presents the full heavy metal mass balance of black water based on literature data with additional control samples analyzed within this study. The contribution of different black water components to the total heavy metal loading of black water is determined and compared with the heavy metal loading of sewage. To argue for targeted sludge reuse regulation, a control parameter is suggested to distinguish black water from sewage. To further promote soil application of black water sludge, the primary origin of heavy metals in the black water components is investigated, and the impact of current agricultural practices on the heavy metal content in the soil/food cycle is discussed.

3.2 Materials and Methods

3.2.1 Heavy metal loading calculations

Heavy metal loading (mg/cap/day) was used as a parameter to draw the heavy metal mass balance of black water, to calculate the contribution of different black water components, and to determine the dietary excretion of heavy metals from the human body. The heavy metal loading of black water components and black water was calculated according to Eq. (3.1)

$$Q = C * DW * P \tag{3.1}$$

where Q is the heavy metal loading (mg/cap/day), C is the heavy metal concentration (mg/gDW), DW is the dry weight of the sample (g/L), and P is the production rate of the sample (L/cap/day or gDM/cap/day) presented in Table 3.1.

Table 3.1: *Production rate of feces, urine, flush water, toilet paper, toilet detergent and concentrated black water*

Sample	Unit	Production rate
Feces	gDM/cap/day	30^1
Urine	L/cap/day	1^2
Flush water	L/cap/day	6^2
Toilet paper	gDM/cap/day	14^3
Toilet detergent	gDM/cap/day	1^*
Concentrated black water	L/cap/day	7.5^2

(1) [92]
(2) [91] (based on vacuum toilet)
(3) [200]
DM=dry matter
* assumed based on consumption of 1 toilet rim block/cap/month

3.2.2 Literature data

Extensive literature study was done to acquire data on the heavy metal concentrations of different black water components and black water, and the heavy metal loading of food (diet). The data was collected from studies conducted in 11 different countries within Europe during the last 30 years. The location, sample size and reference of literature data on feces, urine, flush water and black water is presented in Table 3.2. In this study flush water is considered to be tap water. The location, type of study and reference of literature data on food is presented in Table 3.3. Three different types of studies were included: duplicate diet study, total diet study and direct analysis. In duplicate diet study an ordinary diet is consumed from which a duplicate portion is prepared for analysis. Total diet study, also known as market basket analysis, determines the level of contaminants in various food products and estimates the dietary intake of a population. In direct analysis, randomly selected food products are analysed and the dietary intake is determined.

Table 3.2: *Location, sample size and reference of literature data on feces, urine, flush water and black water*

Sample	Location	Sample size	Reference
Feces	UK	23	[25]
	Belgium	25	[29]
	Sweden	27	[29]
	Sweden	15	[194]
	Sweden	20	[196]
	Sweden	nd	[208]
	Sweden	15	[195]
	Croatia	17	[195]
	LU	40	[11]
Urine	UK	23	[25]
	Italy	nd	[128]
	Sweden	20	[173]
	Sweden	13	[88]
	Germany	14	[162]
	Germany	4000	[90]
	LU	40	[11]
Flush water	Germany	5100	[90]
	Italy	36	[178]
Black water	Sweden	3	[143]
	NL	nd	[91]

Table 3.3: *Location, type of study and reference of literature data on food (diet)*

Location	Type of study	Reference
Belgium	Duplicate Diet study	[24]
NL	Total Diet study	[47]
France	Direct analysis	[13]
France	Total Diet study	[106]
UK	Total Diet study	[223]
UK	Total Diet study	[152]
Denmark	Direct analysis	[99]
Spain	Direct analysis	[15]
Italy	Total Diet study	[189]
Sweden	Total Diet study	[10]

3.2.3 Sample collection

To cross-check the literature data on the heavy metal concentration of black water components and black water, samples were collected at three different locations in the Netherlands all using vacuum toilets (Table 3.4). Additionally, toilet paper and toilet detergent were sampled and analyzed to complete the heavy metal mass balance of black water. Black water sludge was sampled within the study of Tervahauta et al. (2014) [185] during the first 500 days of operation of a black water UASB reactor. After 500 days calcium phosphate granules were formed in the sludge bed. Five different toilet papers were analyzed: two regular, two 100% recycled and one ecological toilet paper made 100% from an agricultural by-product of wheat straw. Three different brands of toilet rim blocks were analyzed as toilet detergent. The wastewater samples were stored at 4°C prior to analysis.

Table 3.4: *Location and sample size of collected samples of black water components, black water, and black water sludge in the Netherlands*

Sample	Location	Sample size
Feces	WUR (NL)	20 (collective sample)
Urine	Wetsus (NL)	2 (collective sample)
Flush water	DESAR (NL)	3
Toilet paper	NL	5
Toilet detergent	NL	3
Black water	DESAR (NL)	7
Black water sludge	Wetsus (NL)	3

NL=the Netherlands
WUR=Wageningen University
DESAR=Decentralized Sanitation and Reuse demonstration site, Sneek
Wetsus=Centre of Excellence for Sustainable Water Technology, Leeuwarden

3.2.4 Sample preparation and analysis

The heavy metal concentration of feces, toilet paper, toilet detergent, black water, and black water sludge was determined from the dry matter fraction, and the heavy metal concentration of urine and flush water was determined from the unfiltered sample. The dry weight was determined by drying the sample at 105°C overnight and by recording the weight. The dry matter fraction was then acid digested using Ethos 1 Advanced Microwave digestion system of Milestone. The dried sample (0.5 g) was placed in a special microwave vessel with 10 mL of nitric acid (68 %). To ensure a complete destruction of the sample, a mixture of nitric acid (2.5 mL) and hydrochloric acid (37%) (7.5 mL) was used for toilet paper, and a mixture of nitric acid (6 mL), sulfuric acid (96%) (2 mL) and milliQ water (1 mL) was used for toilet detergent. The samples were heated in the microwave at 180°C for 25 minutes. The acid digestion was done in duplicate and the relative standard deviation (%RSD) was controlled within 20%. Heavy metals (As, Zn, Cu, Ni, Cd, Pb, Hg and Cr) were then analyzed from the digestate with Inductively Coupled Plasma Optical Emission Spectrometry (ICP–AES) (Perkin Elmer Optima 5300174 DV). Urine and flush water were directly analyzed with ICP–AES after addition of nitric acid to reach acid concentration of 1% to retain heavy metals in solution. The limit of detection (LOD) for ICP–AES was 25 ppb for Cu, Cd, Cr, Zn and Ni, and 250 ppb for As and Pb. The heavy metals below these limits were analyzed with Inductively Coupled Plasma Mass Spectrometry (ICP-MS) in an external lab with LOD of 0.02 ppb for Hg, 0.1 ppb for Cd, 1 ppb for As, Cr, Pb and Ni, 5 ppb for Cu, and 10 ppb for Zn. The ICP–AES/MS analysis was done in duplicate and the %RSD was controlled within 5% and for toilet paper within 7%.

3.2.5 Statistical analysis

Statistical analysis using Statdisk software was used to compare the contribution of different black water components to the total heavy metal loading of black water. Hypothesis test using the sample size, average heavy metal loading and standard deviation was used to evaluate the confidence interval at which a certain black water component is the major contributor in the heavy metal loading of black water. The normality of the data sets was defined according to Hair et al. (1998) [69], and was confirmed normally distributed at a confidence interval of 99% for feces and 95% for urine. However, it should be noted that the normality was calculated using the average values from the literature studies, and the size of the data sets was therefore limited. Furthermore, due to this reason the normality of the flush water data set was not possible to confirm.

3.3 Results

3.3.1 Heavy metal mass balance of black water

Tables 3.5, 3.6 and 3.7 present the heavy metal loading of feces, urine, flush water, toilet paper, toilet detergent and black water according to both literature and experimental data, calculated according to Eq. (3.1). The calculated total heavy metal loading of the black water components is compared with the measured heavy metal loading of black water.

Table 3.5: *Heavy metal loading of feces, urine, flush water, toilet paper, toilet detergent and black water according to literature and experimental data (As, Pb, Cd) (Unit mg/cap/day)*

	As				**Pb**				**Cd**			
Sample	Lit.		Exp.		Lit.		Exp.		Lit.		Exp.	
	Avg	Std	Avg	Std	Avg	Std	Avg	Std	Avg	Std	Avg	Std
Feces	nd	nd	nd	-	0.041	0.030	0.023	-	0.010	0.005	0.010	-
Urine	0.030	0.023	0.037	-	0.011	0.018	0.008	-	0.001	0.001	nd	-
Flush water	0.034	0.024	nd	nd	0.020	0.019	nd	nd	0.001	0.000	nd	nd
Toilet paper	-	-	nd	nd	-	-	0.007	-	-	-	0.0003	-
Detergent	-	-	nd	nd	-	-	nd	nd	-	-	nd	nd
BW												
Calculated	**0.063**	**0.047**	**0.037**	**-**	**0.072**	**0.066**	**0.039**	**-**	**0.012**	**0.006**	**0.010**	**-**
Measured	**nd**	**nd**	**0.015**	**0.006**	**0.076**	**0.083**	**0.068**	**0.034**	**0.007**	**0.006**	**0.009**	**0.004**

Lit.=Literature data
Exp.=Experimental data
Avg=Average
Std=Standard deviation
nd=not detected
- not determined

Table 3.6: *Heavy metal loading of feces, urine, flush water, toilet paper, toilet detergent and black water according to literature and experimental data (Cu, Zn, Cr) (Unit mg/cap/day)*

	Cu				**Zn**				**Cr**			
Sample	Lit.		Exp.		Lit.		Exp.		Lit.		Exp.	
	Avg	Std	Avg	Std	Avg	Std	Avg	Std	Avg	Std	Avg	Std
Feces	1.23	0.233	1.68	-	10.5	0.395	8.53	-	0.020	0.000	0.059	-
Urine	0.054	0.048	0.064	-	0.360	0.503	0.266	-	0.005	0.005	nd	-
Flush water	0.559	0.743	1.35	1.11	1.97	1.90	1.47	0.891	0.003	0.000	nd	nd
Toilet paper	-	-	0.107	0.013	-	-	0.247	0.069	-	-	nd	nd
Detergent	-	-	nd	nd	-	-	2.1*10-5	1.2*10-6	-	-	nd	nd
BW												
Calculated	**1.84**	**1.02**	**3.20**	**1.12**	**12.8**	**2.80**	**10.5**	**0.960**	**0.028**	**0.005**	**0.059**	**-**
Measured	**1.26**	**0.443**	**4.68**	**2.51**	**5.36**	**2.00**	**14.1**	**4.21**	**0.058**	**0.050**	**3.82**	**4.04**

Lit.=Literature data
Exp.=Experimental data
Avg=Average
Std=Standard deviation
nd=not detected
- not determined

Table 3.7: *Heavy metal loading of feces, urine, flush water, toilet paper, toilet detergent and black water according to literature and experimental data (Ni, Hg) (Unit mg/cap/day)*

	Ni				**Hg**			
Sample	Lit.		Exp.		Lit.		Exp.	
	Avg	Std	Avg	Std	Avg	Std	Avg	Std
Feces	0.116	0.073	0.146	-	0.013	0.006	nd	-
Urine	0.016	0.017	nd	-	0.006	0.005	nd	-
Flush water	0.019	0.033	0.814	0.359	nd	nd	nd	nd
Toilet paper	-	-	0.005	-	-	-	nd	nd
Detergent	-	-	nd	nd	-	-	nd	nd
BW								
Calculated	**0.150**	**0.122**	**0.965**	**0.359**	**0.018**	**0.012**	**nd**	**nd**
Measured	**0.119**	**0.071**	**2.28**	**2.33**	**0.005**	**nd**	**0.0003**	**0.0002**

Lit.=Literature data
Exp.=Experimental data
Avg=Average
Std=Standard deviation
nd=not detected
- not determined

The literature and experimental data on the heavy metal loading of feces are similar, and the relatively low standard deviations indicate consistent feces composition, also realized in regions outside Europe [130, 161]. The As content of feces is below the detection limits, and Hg is only detected in the literature data. The differences between the literature and experimental data on Hg is possibly due to different local conditions and demographic aspects [170].

The literature and experimental data on the heavy metal loading of urine are similar for As, Pb, Cu and Zn. Possibly due to the different locations and sample sizes, Cd, Cr, Ni and Hg are only detected in the literature data. The high standard deviations indicate highly variable urine composition. In the study of Kuntke (2013) [95], the amount of organic compounds in urine was shown to strongly correlate with the dilution of urine. However, as the inorganic compounds in urine did not show as strong correlation with the dilution, the high standard deviations for heavy metals in urine cannot be explained by it. Instead, the highly variable composition of tap water used for drinking could influence the variation in the urine composition.

The type of water source, the regional geological conditions and the piping materials influence the changes in tap water composition [90]. The heavy metal loading of tap water in the regional pumping station providing water to the DESAR demonstration site in Sneek (NL) (Cu and Zn $\leq$ 0.03 and Ni $\leq$ 0.006 mg/cap/day in Spannenburg pumping station [211]) is significantly lower compared to the loading of tap water at the demonstration site (Cu 1.35, Zn 1.47 and Ni 0.81 mg/cap/day), indicating an input of heavy metals to tap water from the distribution network. The strong influence of piping materials on the Cd, Cu, Zn and Pb loading in tap water has also been shown in other studies [31, 90].

Due to the lack of recent literature data, only experimental data are presented for toilet paper and toilet detergent. Toilet paper contains mostly Zn and Cu, and small amounts of Pb, Cd and Ni. Similar Cd and Pb content of recycled toilet paper is presented in the study of Storr-Hansen and Rastogi (1988) [175], while no Hg is detected in toilet paper in this study, and might be due to the overall decrease in Hg levels in the environment [206]. The Cu and Zn loading from the wheat straw toilet paper (Cu 0.09 and Zn 0.22 mg/cap/day) is slightly lower compared to the wood derived toilet paper (Cu 0.11 and Zn 0.25 mg/cap/day). However, the Cd, Pb and Ni loading from the wheat straw toilet paper (Cd 0.002, Pb 0.03 and Ni 0.02 mg/cap/day) is around 10-fold higher compared to the wood derived toilet paper

(Cd 0.0003, Pb 0.007 and Ni 0.005 mg/cap/day), and has an additional Cr loading of 0.04 mg/cap/day. The use of wheat straw toilet paper is therefore not considered in this study. Only small amount of Zn is detected in toilet detergent, contributing less than 0.0005% to the calculated total heavy metal loading of black water. Similarly, the heavy metal content of most household detergents is found to be low [31]. The low standard deviations indicate consistent composition of toilet paper and toilet detergent.

The measured heavy metal loading of black water is either lower or similar to the calculated total heavy metal loading of black water components, indicating absence of external heavy metal input to toilets such as household chemicals. However, the measured Cr and Ni loading of black water according to experimental data is higher compared to the calculated loading, and could be explained by a temporary input of household chemicals to the toilets. The high standard deviations of Cr and Ni in the experimental data could be related to the use of Cr and Ni in metal finishing processes for stainless steel used in pipes, pumps and fittings, and the wearing off of the metal coating over time [155]. The overall high standard deviations of the measured heavy metal loadings of black water indicate highly variable black water composition that can originate from regional and seasonal changes in the household activities. Nevertheless, the heavy metal mass balance of black water is closed with the exception of As that is correlating with local geochemical conditions [213].

3.3.2 Heavy metal contribution from black water components

Based on the literature data on average heavy metal loadings of black water (Table 3.5, 3.6 and 3.7), feces contributes major part to the Pb, Cd, Cu, Zn, Cr, Ni and Hg loading of black water (52%, 84%, 63%, 80%, 72%, 75% and 69%, respectively). Flush water has a high contribution to the Pb (25%) and Cu (29%) content of black water, and can be related to the use of Pb and Cu piping materials [31, 90]. The As loading of black water originates from flush water (53%) and urine (47%), and the Hg loading originates from feces (69%) and urine (31%). The contribution of toilet paper to the Pb, Cu, Zn, Cd and Ni content of black water stays within 10%. According to the statistical analysis, the contribution of urine to the As loading of black water is either lower or equal to the contribution of flush water at a confidence interval of 95%. The contribution of feces to the Pb, Cd, Cu, Zn, Cr and Ni loading of black water is higher than the contribution of flush water at a confidence interval of 95%.

Feces can be therefore considered as the main contributor to the heavy metal loading of black water with the exception of As.

In the study of Koch and Rotard (2001) [90], tap water is found to be one of the most significant contributors to the heavy metal loading of municipal sewage, in particular Cu and Zn, while feces accounts for less than 10% of the heavy metal loading and urine is considered to be a negligible source. As concentrated black water contains a significantly smaller fraction of tap water compared to sewage, the contribution of heavy metals from tap water decreases, while increasing the heavy metal contribution from feces and urine. The vacuum collection of black water therefore creates a wastewater stream characterized by human originated content (feces and urine) rather than infrastructure originated content (tap water).

3.3.3 Origin of heavy metals in feces and urine

The absorption and excretion of heavy metals in the human body can be defined by the exposure media, major uptake pathways, transport and distribution, and major excretory pathways [7]. The exposure media can be divided to air, water, food and medicine, the major uptake pathways can be divided to skin, respiratory tract and gastrointestinal tract, the transport and distribution is done via blood and organs (in particular liver and kidney), and the major excretory pathways can be divided to sweat, hair, urine and feces. To investigate the correlation between the major heavy metal uptake and excretory pathways, the average heavy metal loading of food, feces and urine according to literature data are used to compare the calculated and theoretical dietary excretion of heavy metals from the human body (Table 3.8). The calculated dietary excretion (%) was determined according to Eq. (3.2)

$$Dietary\ excretion = \frac{Q_{feces} + Q_{urine}}{Q_{food}} * 100\% \tag{3.2}$$

where Q_{feces} and Q_{urine} are the average heavy metal loadings (mg/cap/day) of feces and urine according to literature data presented in Table 3.5, 3.6 and 3.7, and Q_{food} is the average heavy metal loading (mg/cap/day) of food presented in Table 3.8.

Table 3.8: *Literature data on the average heavy metal loading of food (mg/cap/day) and dietary excretion of heavy metals from the human body (%)*

Element	Food*		Dietary excretion (%)	
	Avg	Std	Calculated	Theoretical
As	0.106	0.175	28	70[1]
Pb	0.034	0.029	153	90[2]
Cd	0.014	0.006	78	95[3]
Cu	1.25	0.179	103	25–45[4]
Zn	11.3	2.35	96	60–80[5]
Cr	0.128	0.118	19	98[6]
Ni	0.168	0.090	79	99[7]
Hg	0.005	0.003	405	5–85[8]

* references presented in Table 3.3
(1) [154]
(2) [30]
(3) [214]
(4) [180]
(5) [179]
(6) [136]
(7) [67]
(8) [127]
Avg=Average
Std=Standard deviation

The different dietary excretion fractions are due to the different functions of heavy metals in the human body, and depends on the form and solubility of the metal compound ingested. Cu and Zn are essential trace elements, therefore, their excretion from the human body is generally lower compared to As, Pb, Cd, Cr, Ni, Hg that either have no significant function in the metabolic system or are toxic already at low levels. The calculated excretion of Pb, Cu, Zn and Hg is higher compared to the theoretical excretion, and external sources are therefore considered to contribute to the load of these heavy metals in feces and urine. External sources can be drinking water, medicine, mineral supplements and air pollution. In addition, Hg is known to originate from dental amalgam [66, 170]. The calculated excretion of As, Cd, Cr and Ni is lower compared to the theoretical excretion, and no external sources are therefore considered to contribute to the load of these heavy metals in feces and urine. The differences in geochemical conditions and agricultural practices in different regions further influence the variation in the heavy metal load in food, and thus the calculated dietary excretion of heavy metals. The data in Table 3.8 show that the heavy metals in feces and urine primarily originate from dietary sources, except for Hg.

In the study of Rose et al. (2010) [152], the contribution of different food groups to the dietary exposure of heavy metals is determined. The main contributors to the Pb, Cd, Cu, Cr, Ni and Hg loading of the diet are bread, cereals, fruits, potatoes and vegetables (57%, 73%, 61%, 46%, 41%, and 29%, respectively). Meat and dairy products contribute the largest part to the Zn loading (56%) and fish contributes the largest part to the As loading (88%) of the diet. Other food groups contributing to the heavy metal loading of the diet are sugars and preserves (1–16%), and beverages (2–21%). As majority of the heavy metals in the diet originate from agricultural products (bread and cereals, fruit and vegetables, meat and dairy), the dietary exposure of heavy metals can be linked to agricultural practices.

3.4 Discussion

3.4.1 Closing the agricultural heavy metal cycle

According to Nicholson et al. (2003) [133], the main sources of heavy metals entering agricultural soil in England and Wales can be ascribed to atmospheric deposition, livestock manures, sewage sludge and inorganic fertilizers. The heavy metal input rates (g/ha/year) from the different sources are compared by assuming an application rate equivalent to 250 kgN/ha/year for sewage sludge and livestock manures. Based on this comparison, the soil application of sewage sludge generates the highest input rates of all heavy metals to agricultural soil. Major part of the Pb and Hg input is further contributed by atmospheric deposition, originating from industrial activities such as energy production, mining and waste incineration. Heavy metals in livestock manures originate from the diets, drinking water and housing of the livestock, and contribute major part to the Zn and Cu input. Among the inorganic fertilizers, phosphate fertilizer has a significant heavy metal content that originates from phosphate rock, and contributes major part to the Cd and Cr input.

Although the soil application of sewage sludge promotes closing of the carbon and nutrient cycle, the high input rates of heavy metals to agricultural soil poses a serious threat to the soil quality. Major part of the heavy metals in sewage sludge originates from industrial effluents and surface run-offs, and yet many sources are unknown [90, 170]. Given the new developments in waste management and sanitation, a new sludge reuse regulation is required following a source-oriented approach to allow feces and urine to return to the soil/food cycle without external heavy metal inputs. As black water sludge is predominantly human originated (feces and urine), it should be differentiated from sewage sludge in the sludge reuse regulation to promote carbon and nutrient recovery while improving the soil quality. This could be done by defining a control parameter, such as the Hg and Pb content that is significantly higher in sewage sludge compared to black water sludge (from 50- to 200-fold) (Table 3.9), to distinguish these two streams in the sludge reuse regulation.

Table 3.9: *Heavy metal content of black water sludge, sewage sludge, cow manure and phosphate fertilizer (Unit mg/kgP)*

Element	BW sludge*	Sewage sludge[1]	Cow manure[2]	P-fertilizer[3]
As	12	300	nd	33
Cd	13	39	33	91
Cr	731	1268	1145	1245
Cu	3720	12701	14397	207
Hg	0.12	23	nd	0.7
Ni	466	1025	1472	202
Pb	69	3519	695	154
Zn	13919	31166	25947	1923

* measured in this study
(1) [27]
(2) [202]
(3) [149]
nd=not detected

To decrease the amount of heavy metals in feces and urine, external sources of heavy metals entering agricultural soil and sub-sequently the food products need to be reduced. Including manure and artificial fertilizers in the sludge reuse regulation is therefore recommended. One of these examples is the ongoing plan to regulate the Cd content of phosphate fertilizers in the European market [135]. For the sludge reuse regulation to reflect more the application rate of inorganic and organic fertilizers, the maximum permissible heavy metal content should be based on the phosphorus content of these products. As black water sludge has a significantly lower heavy metal content compared to cow manure, and a lower As, Cd, Cr, Hg and Pb content compared to phosphate fertilizers per kg phosphorus (Table 3.9), black water sludge should replace part of the manure and phosphate fertilizers applied in agricultural land, provided that the bio-availability of phosphate in black water sludge is sufficient. Zeeman (2012) [224] calculates that at full recovery phosphorus in black water and kitchen refuse can replace 25% of the present global artificial phosphate fertilizer need. As manure represents a significant stream of carbon and nutrients, alternative ways to decrease the heavy metal content, such as heavy metal regulation of the livestock feed, should be ultimately implemented for manure to be utilized in agriculture. To minimize the risk related to pathogens when applying wastewater sludge in agricultural soil, pasteurisation of the sludge at 70°C should be performed [216]. For this reason a hyper-thermophilic treatment of sludge could be implemented [114]. Hyper-thermophilic treatment can be applied energy efficiently on black water that is collected with highly water efficient toilets [224].

3.4.2 Toxicity of heavy metals

The regulations on heavy metals entering agricultural soil need to take into account the greatly varying toxicity of the metals and the characteristics of the soil. As Cu and Zn are essential trace elements, they become toxic only at high levels of exposure, while Ni, Cr, Cd, Pb and Hg have no essential role in the metabolic system and are toxic already at low concentrations (Cd, Pb and Hg), or can be tolerated at medium to high concentrations (Ni and Cr) [139, 203]. Within the EU the maximum permissible concentrations of Zn and Pb in the soil are the same (300 mg/kgDW), while in the U.S. the permissible concentration of Pb is significantly lower compared to Zn (150 mgPb/kgDW and 1400 mgZn/kgDW), reflecting more the different toxicity of the heavy metals (Renner, 2000). The EU Directive 86/278/EEC is currently under revision and a study is launched to examine the environmental, economic, social and health impacts of present sewage sludge reuse practices in agricultural soil [55]. In order to protect the soil quality, a new heavy metal regulation within the EU is needed that differentiates black water sludge from sewage sludge, promoting the reuse of black water sludge in agriculture.

3.5 Conclusions

- According to the heavy metal mass balance, black water does not contain external heavy metal sources, such as household chemicals.
- Feces is the main contributor to the heavy metal loading of vacuum collected black water (52%–84%), while in sewage the contribution of feces is less than 10%.
- To distinguish black water from sewage in the sludge reuse regulation, a control parameter should be implemented, such as the Hg and Pb content that is significantly higher in sewage sludge compared to black water sludge (from 50- to 200-fold).
- The heavy metals in feces and urine are primarily from dietary sources, and promotion of the soil application of black water sludge over livestock manure and artificial fertilizers could further reduce the heavy metal content in the soil/food cycle.

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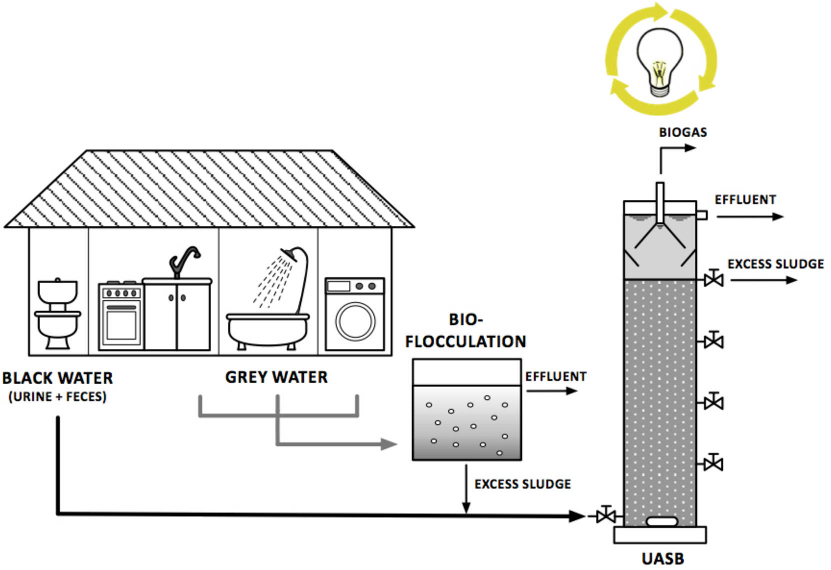
Chapter 4

Improved energy recovery by anaerobic grey water sludge treatment with black water



Abstract

This study presents the potential of combining anaerobic grey water sludge treatment with black water in an upflow anaerobic sludge blanket (UASB) reactor to improve energy recovery within source-separated sanitation concepts. Black water and the mixture of black water and grey water sludge were compared in terms of biochemical methane potential (BMP), UASB reactor performance, COD mass balance and methanization. Grey water sludge treatment with black water increased the energy recovery by 23% in the UASB reactor compared to black water treatment. The increase in the energy recovery can cover the increased heat demand of the UASB reactor and the electricity demand of the grey water bioflocculation system with a surplus of 0.7 kWh/cap/y electricity and 14 MJ/cap/y heat. However, grey water sludge introduced more heavy metals in the excess sludge of the UASB reactor, and might therefore hinder its soil application.

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4.1 Introduction

Biogas as a renewable energy source is increasing its energy market share with the enforced regulation of organic waste treatment and recycling in the European Union (EU) [77]. Co-digestion is a well-known concept for improving the biogas yield from different wastewater streams and biosolids due to positive synergisms in the microbiota, supply of nutrients, dilution of inhibitive compounds, and improved moisture content of the feed [121]. Several studies have investigated the co-digestion of different substrates such as sewage sludge and grease trap sludge [38, 116], cattle slurry and cheese whey [33], potato processing water and glycerol [117], and a number of different animal manures and energy crops, also mentioned in the White Paper of renewable energy sources (RES) of the EU-Commission from 1997 [34].

Anaerobic treatment of source-separated domestic wastewater is recognized as the core technology to improve energy recovery from domestic wastewater [227]. Concentrated black water (toilet water) and kitchen refuse has been traditionally considered as the main source for energy recovery in the decentralized sanitation and reuse (DESAR) concept [91]. Co-digestion of kitchen refuse with black water has shown improved methane production in both accumulation systems [93] and UASB septic tanks [115]. Grey water originating from washing activities in the household, however, contributes a significant fraction of the organic load present in domestic wastewater. Currently, this fraction is lost either by using a constructed wetland or a sequencing batch reactor (SBR) [1], where the organic matter is oxidized instead of utilizing it as an energy source. Alternatively, the organic fraction can be concentrated in a bioflocculation unit, such as the membrane bioreactor (MBR) or the A-trap of the AB-process [16], and sub-sequently treated with black water in an up-flow anaerobic sludge blanket (UASB) reactor for improved energy recovery. In the study of Hernández Leal et al. (2010) [74], grey water was bioflocculated in a MBR, and anaerobic batch experiments on the produced sludge indicated a high biochemical methane potential (BMP) (88%).

No studies, however, have experimentally investigated grey water sludge treatment with black water in both batch and continuous UASB reactor experiments. This study presents the potential of grey water sludge treatment with black water in batch experiments by determining the BMP of black water, grey water sludge from an A-trap and their mixture. Continuous experiments are further conducted to compare the reactor performance, COD mass balance and methanization of two UASB reactors: one operated on black water and one on the mixture. In addition, the influence of grey water sludge addition on the quality of UASB reactor excess sludge in terms of heavy metals is examined to evaluate its application in soil improvement.

4.2 Materials and Methods

4.2.1 Grey water sludge and black water source

Every two weeks grey water sludge and concentrated black water (vacuum collected) were collected from the 32 houses in the DESAR demonstration site in Sneek, the Netherlands [74]. Grey water sludge was collected from a storage tank connected to the bottom of the settling tank of the A-trap and black water was collected from a buffer tank. Grey water sludge and black water was transported to the experimental hall in Leeuwarden and stored in a cold room at 4°C before feeding to the reactors.

4.2.2 Experimental setup of the UASB reactors

In this study two 50 L UASB reactors were operated at 25°C for 490 d on vacuum collected black water and 498 d on the mixture of black water and grey water sludge (Figure 4.1). The COD based mixture ratio of 5:1 (black water:grey water sludge) was according to the actual COD loadings of these two streams at the DESAR demonstration site. Steady state was assumed after 90 d of operation with stable methane production. The reactors were inoculated with 20 L of anaerobic sludge (9.7 gVSS/L) from an UASB reactor operated on vacuum collected black water at 25°C. The details of the reactor are described in the study of de Graaff et al. (2010) [42].

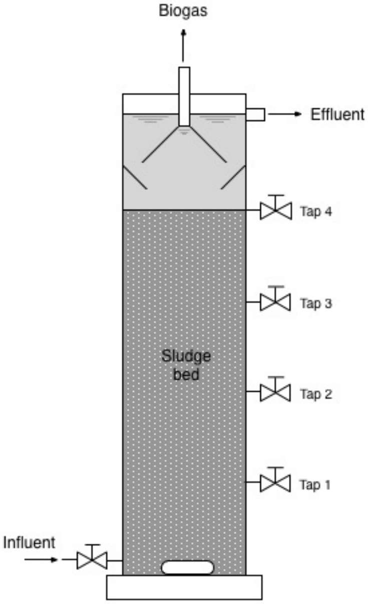

Figure 4.1: *UASB reactor configuration*

4.2.3 Analyses and measurements

Influent and effluent samples (0.5–1 L) (36 samples) were collected weekly and analyzed right after collection for TSS, VSS, COD_{total}, $COD_{suspended}$, $COD_{colloidal}$, $COD_{soluble}$, Total Nitrogen (TN), Total Phosphorus (TP), Anionic Surfactants (AS), total ammonia nitrogen (NH_4–N), Volatile Fatty Acid (VFA) (acetic acid, propionic acid and butyric acid), anions ($PO_4{}^{3-}$, Cl^-, $SO_4{}^{2-}$, NO^{2-} and NO^{3-}), soluble elemental phosphorus ($TP_{soluble}$) and Inorganic carbon (IC). The details of the sample collection and analysis are described in the study of de Graaff et al. (2010) [42]. The AS concentration was determined with cuvette tests based on the MBAS standard method [6]. Biological Oxygen Demand (BOD) was determined from the UASB reactor effluent (4 samples). Biogas composition was analyzed from 9 samples with gas chromatography (Shimadzu GC-2010 Gas Chromatograph containing GS-Q (CO_2) and HP molsieve (O_2, N_2, H_2S and CH_4) columns. Excess sludge (14 samples) and sludge bed samples taken from tap 1, 2, 3 and 4 (3 times 4 samples) were analyzed for COD_{total}, TSS and VSS. The hydraulic retention time (HRT), sludge retention time (SRT) and the COD mass balance were calculated as described in the study of de Graaff et al. (2010) [42].

Total lipid, protein and carbohydrate content was analyzed from the UASB reactor influent (4 samples) and grey water sludge (1 sample). Total lipid was determined using the Bligh-Dyer extraction method from acidified samples [14], and measuring the lipids gravimetrically after the solvent was evaporated at 80°C [6]. Total protein was determined from the difference between the corresponding TN and ammonia nitrogen concentrations, and dividing the difference by 0.16 [83]. 1 g protein (assumed as $(C_4H_{6.1}O_{1.2}N)_x$) was considered to equal to 1.5 g COD [129] and the remaining COD was termed as carbohydrates.

4.2.4 Batch experiments

Biochemical methane potential (BMP), calculated as a percentage of the influent COD converted to methane, was determined from black water, grey water sludge and the mixture. The experiment was done in triplicate in 500 mL glass bottles with Oxitop heads according to the study of Kujawa (2005) [91]. The inoculum sludge used in the experiment was anaerobic sludge from a municipal wastewater treatment plant in Leeuwarden (20 gVSS/L). The bottles were placed on a shaker and incubated at 35°C for 60 d. The pH stayed within the range of 7.5-8.3 during the experiments.

4.2.5 Statistical analysis

Statistical analysis of the data was done using the hypothesis testing of Statdisk. The normality of the data sets was defined according to Hair et al. (1998) [69] and was confirmed normally distributed at a confidence interval of 95%.

4.2.6 Energy recovery calculations

The methane production (L/cap/d) in the UASB reactors was calculated by determining the COD load of black water and grey water sludge (86 gCOD/cap/d for black water and 15 gCOD/cap/d for grey water sludge according to the production at the DESAR demonstration site [45]), and using the fraction of incoming COD converted to methane from the COD mass balance of the UASB reactors. The volume of the produced methane was calculated using a theoretical methane production of 0.35

L/gCOD (STP) and the primary energy production from methane was calculated using the volume of methane and the calorific value of methane (35.8 MJ/m^3) [181]. The increased heat demand of the MIX-UASB reactor was due to the heating of the additional infuent stream of grey water sludge and the increased heat loss through the reactor walls of the larger reactor. The heat demand was calculated according to Tervahauta et al. (2013) [183] by using grey water sludge production of 1.0 L/cap/d and temperature of 19°C (annual average) [45], operational temperature of 25°C for the UASB reactor, and ambient temperature at the DESAR demonstration site of 19°C (annual average) [45].

4.2.7 Heavy metal analysis

To assess the influence of grey water sludge addition on the quality of UASB reactor excess sludge, heavy metals were analyzed in 3 samples of grey water sludge, and the influent and excess sludge of the UASB reactors. Prior to the analysis, samples were dried at 105°C overnight and acid digested using the Ethos 1 Advanced Microwave digestion system of Milestone. Dried sample (0.5 g) was put into a special microwave vessel with 10 mL of nitric acid (68%). The samples were heated in the microwave at 180°C for 25 minutes. After cooling down, the samples were diluted to reach an acid concentration of 1%. The acid destruction was done in duplicate and the Relative Standard Deviation (%RSD) was controlled within 20%. The samples were analyzed with Inductively Coupled Plasma-Atomic Emission Spectroscopy (ICP-AES) for arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), mercury (Hg), nickel (Ni), lead (Pb), and zinc (Zn). The limit of detection (LOD) for ICP–AES was 25 ppb for Cu, Cd, Cr, Zn and Ni, and 250 ppb for As and Pb. The heavy metals below these limits were analyzed with Inductively Coupled Plasma Mass Spectrometry (ICP-MS) in an external lab with LOD of 0.02 ppb for Hg, 0.1 ppb for Cd, 1 ppb for As, Cr, Pb and Ni, 5 ppb for Cu, and 10 ppb for Zn. The ICP–AES/MS analysis was done in duplicate and the %RSD was controlled within 5%.

4.3 Results and Discussion

4.3.1 Energy recovery

To determine energy recovery in grey water sludge treatment with black water, two UASB reactors, one operated on black water and one on the mixture (5 BW : 1 GW-S), are examined for the COD mass balance and methanization. Figure 4.2 presents the COD mass balance of the UASB reactors over the total period of operation of 490 d for the BW-UASB reactor and 498 d for the MIX-UASB reactor. The total amount of COD fed to the reactor during this time is 21 kg of which 3% is inoculum sludge for the BW-UASB reactor and 30 kg of which 1% is inoculum sludge for the MIX-UASB reactor. *Methane* is the amount of CH_4–COD produced in the reactor, *Effluent* is the amount of total COD discharged with effluent, *Sludge wasted* is the amount of COD wasted as excess sludge including sludge bed samples, and *Sludge reactor* is the amount of COD accumulated in the sludge bed. The difference between the total incoming COD and total outgoing COD is 8% for the BW-UASB reactor and 4% for the MIX-UASB reactor, and this fraction can be explained by errors in sampling and analyses. The amount of CH_4–COD produced in the MIX-UASB reactor (63%) is higher compared to the BW-UASB reactor (60%). Similarly, the BMP determined in the batch experiments is higher for the mixture (88%) compared to black water (61%) (Table 4.1). The BMP of the mixture is higher than the calculated one based on the mixture ratio of 5:1 (BW:GW-S) (65%), and could be due to synergistic effects of micro-organisms increasing the biodegradable fraction in black water as a result of grey water sludge addition [171, 3]. The mechanisms behind these processes, however, are not yet known.

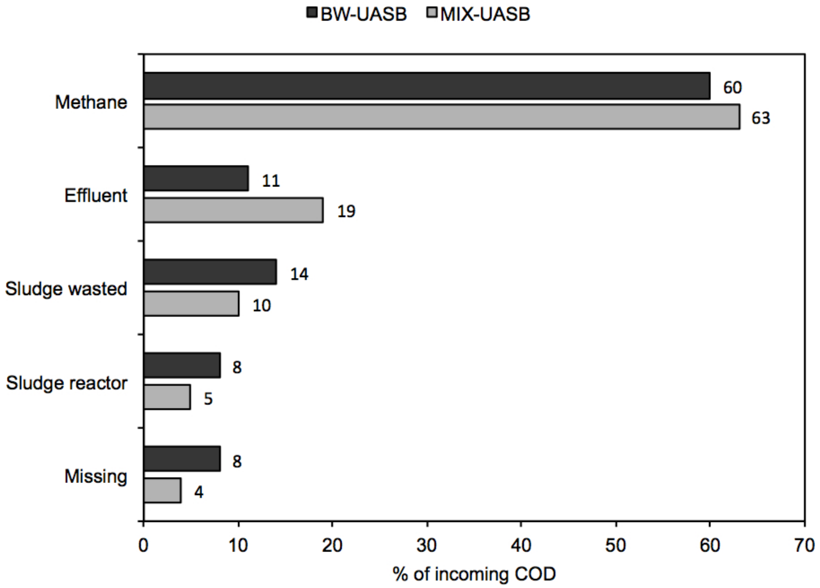

Figure 4.2: *COD mass balance of the UASB reactors over the total period of operation (490 d for BW-UASB reactor and 498 d for MIX-UASB reactor)*

Table 4.1: *Sample composition and BMP in the batch experiments*

Parameter	Unit	Black water	Grey water sludge	MIX (5 BW : 1 GW-S)
COD_{total}	g/L	8.5	25	11
AS	mg/L	189	987	309
BMP	%	61 (10)	92 (0)	88 (3)

( ) standard deviation

Surfactants and lipids present in the sample can also influence the digestion process. Surfactants are known to inhibit methanogens [60], and the high AS content of grey water sludge and the MIX-UASB reactor influent (523 and 328 mg/L, respectively) compared to the BW-UASB reactor influent (189 mg/L) (Table 4.2) could therefore result in a decreased methane production. However, as lipids have a higher anaerobic biodegradability compared to carbohydrates and proteins [5], the high lipid content of grey water sludge and the MIX-UASB reactor influent (74 and 59 wt%, respectively) compared to the BW-UASB reactor influent (20 wt%) (Table 4.2) can increase the methane production in the MIX-UASB reactor.

Table 4.2: *Surfactants and organic components in the influent of the UASB reactors and the grey water sludge added to the MIX-UASB reactor*

Parameter	Unit	BW-UASB	MIX-UASB (5 BW : 1 GW-S)	
		Influent	Influent	GW sludge
AS	mg/L	189 (52)	328 (259)	523 (383)
COD_{total}	gCOD/L	7.1 (1.1)	17 (3.9)	63
Lipid	gCOD/L	1.4 (0.26)	10 (1.9)	47
	wt%	20	59	74
Protein	gCOD/L	4.2 (0.26)	5.6 (2.0)	10
	wt%	58	33	15
Carbohydrate	gCOD/L	1.5 (0.76)	0.8 (1.0)	6.7
	wt%	22	4.7	11

( ) standard deviation

The amount of COD discharged with effluent is higher in the MIX-UASB reactor (19%) compared to the BW-UASB reactor (11%), and might be due to an occational washout of sludge to the effluent. The amount of COD in the wasted sludge and accumulated in the sludge bed are lower in the MIX-UASB reactor (10% and 5%, respectively) compared to the BW-UASB reactor (14% and 8%, respectively), and can be attributed to the higher methane production and the occasional loss of sludge with the effluent in the MIX-UASB reactor.

The two UASB reactors are operated with the aim of applying the same conditions for comparing their performance. Table 4.3 presents the operational conditions and methanization of the UASB reactors at steady state defined as the period of stable methane production after 90 d of start up. The HRT is slightly higher in the MIX-UASB reactor, but does not represent a significant difference at a confidence interval of 95%. The loading rate, however, is higher in the MIX-UASB reactor (1.2 compared to 0.9 kgCOD/m^3/d) due to fluctuations in the influent COD concentration resulting from the grey water sludge addition. Nevertheless, the operational conditions in both UASB reactors are considered comparable. The sludge bed in both UASB reactors is compact and well developed, resulting in a similar sludge concentration and SRT.

The methanization in the UASB reactor is calculated by dividing the total amount of methane produced with the difference between the accumulative influent and effluent COD_{total}. The methanization in the UASB reactor is 5% higher with grey water sludge treatment. Calculated based on the applied HRT, black water production of 5 L/cap/d and grey water sludge production of 1.0 L/cap/d [45], the volume of the reactor at full scale would be 46 L/cap for the BW-UASB reactor and 60 L/cap for the MIX-UASB reactor, resulting in 14 L/cap higher reactor volume for grey water treatment with black water.

Table 4.3: *Operational conditions and methanization of the UASB reactors at steady state*

Parameter	Unit	BW-UASB	MIX-UASB
Temperature	°C	25	25
Loading rate	$kgCOD/m^3/d$	0.9 (0.2)	1.2 (0.5)
HRT	d	9.3 (0.9)	10 (2.2)
SRT	d	138 (73)	131 (90)
Sludge concentration	$gVSS/L_{reactor}$	15 (2.4)	15 (2.9)
Methanization	%	69	74

( ) standard deviation

Grey water sludge treatment with black water increases the energy recovery in source-separated sanitation concepts by introducing an additional organic fraction, and by increasing the methanization in the UASB reactor. The methane production in the MIX-UASB reactor (22.2 L/cap/d) is higher compared to the BW-UASB reactor (18.1 L/cap/d), representing an increase of 23%. This increase is lower compared to the theoretical one calculated in the study of Hernández Leal et al. (2010) [74] (73%), and could be due to the MBR used to bio-flocculate grey water producing a higher loading of grey water sludge of 40 compared to 29 gCOD/cap/d in this study. In the study of Tervahauta et al. (2013) [183], however, the calculated increase in the energy recovery of 28% is similar to this study using an A-trap to bio-flocculate grey water. The increased energy recovery in the MIX-UASB reactor is equivalent to 55 MJ/cap/y primary energy and can cover the increased heat demand of the MIX-UASB reactor of 27 MJ/cap/y with a surplus of 28 MJ/cap/y. Considering an efficiency of 85% of combined heat and power generators, of which 40% is electricity and 60% heat, this primary energy can produce 2.7 kWh/cap/y that covers the electricity demand of the A-trap (2 kWh/cap/y) [183]. Grey water sludge treatment with black water produces an amount of energy that can cover the increased heat demand of the UASB reactor and the electricity demand of the A-trap with a surplus of 0.7 kWh/cap/y electricity and 14 MJ/cap/y heat.

4.3.2 Effluent quality

To investigate the influence of grey water sludge treatment with black water on the effluent quality of the UASB reactor, the reactor performance and effluent quality of the two reactors are compared. Table 4.4 presents the removal efficiencies and characteristics of the influent and effluent of the UASB reactors at steady state, and characteristics of the grey water sludge added to the MIX-UASB reactor. The effluent concentrations of COD_{total}, $\mathrm{COD}_{suspended}$, $\mathrm{COD}_{colloidal}$, $\mathrm{COD}_{soluble}$ and TN are similar in both UASB reactors at a confidence interval of 95%. Furthermore, the effluent concentrations of VFA, TP and BOD_5 can be considered similar in both UASB reactor within the standard deviations. Grey water sludge treatment with black water, therefore, does not deteriorate the effluent quality of the UASB reactor.

Table 4.4: *Removal efficiencies and characteristics of the influent and effluent of the UASB reactors at steady state, and characteristics of the grey water sludge added to the MIX-UASB reactor*

Parameter	Unit	BW-UASB Influent	Effluent	Rem. (%)	MIX-UASB GW sludge	Influent	Effluent	Rem. (%)
pH	-	8.1 (0.23)	7.8 (0.51)	-	7.2 (1.3)	8.1 (0.12)	7.3 (0.18)	-
COD_{tot}	gCOD/L	11 (4.1)	1.1 (0.36)	90	15 (13)	12 (3.9)	1.3 (0.66)	88
COD_{sus}	gCOD/L	6.4 (2.9)	0.23 (0.26)	96	15 (12)	8.5 (3.0)	0.25 (0.21)	96
COD_{col}	gCOD/L	1.2 (0.63)	0.28 (0.13)	77	0.39 (0.37)	0.84 (0.42)	0.30 (0.20)	64
COD_{sol}	gCOD/L	2.9 (0.94)	0.57 (0.075)	80	1.1 (0.68)	1.5 (0.43)	0.53 (0.19)	65
VFA	gCOD/L	0.99 (1.0)	0.020 (0.055)	-	0.85 (0.52)	0.25 (0.15)	0.039 (0.069)	-
TN	gN/L	1.2 (0.22)	1.1 (0.12)	8	0.46 (0.29)	1.3 (0.22)	1.1 (0.21)	15
TP	gP/L	0.19 (0.085)	0.090 (0.034)	53	0.12 (0.07)	0.28 (0.10)	0.14 (0.081)	50
BOD_5	g/L	-	0.16 (0.030)	-	-	-	0.20 (0.050)	-

( ) standard deviation
- not determined

4.3.3 Excess sludge quality

In addition to increased energy recovery by grey water sludge treatment, soil application of black water sludge is a prerequisite for maximum carbon recovery. To investigate the influence of grey water sludge treatment on the UASB excess sludge quality, the heavy metal content (mg/kgDW) of grey water sludge, and the influent and excess sludge of the UASB reactors are determined and compared with the Dutch sludge reuse guidelines (Table 4.5). The heavy metal content of grey water sludge is higher compared to black water (BW-UASB reactor influent) with the exception of Cr, Hg and Ni, resulting in a higher heavy metal content of MIX-UASB reactor influent. The reuse of black water sludge is currently prohibited in the Netherlands due to the elevated Cu and Zn concentrations. The excess sludge of the MIX-UASB reactor has a higher heavy metal content compared to the BW-UASB reactor, but the heavy metals are below the sludge reuse guidelines with the exception of Cu, Zn and Ni. Grey water sludge contributes 36% (Cu), 32% (Zn) and 19% (Ni) to the heavy metal input in the UASB reactor on solid matter basis. Since grey water sludge contributes only 23% to the input of solid matter in the reactor, grey water sludge addition increases the heavy metal concentration in the excess sludge. Furthermore, as heavy metals in black water are primarily human originated (feces and urine) and belong to the soil/food cycle [184], the introduction of external heavy metal sources by grey water sludge addition deteriorates the excess sludge quality, and might therefore hinder its soil application.

Table 4.5: *Heavy metal content (mg/kgDW) of grey water sludge, influent and excess sludge of the UASB reactors, and Dutch sludge reuse guidelines*

Element	BW-UASB		MIX-UASB			Dutch sludge
	Influent	Excess sludge	GW sludge	Influent	Excess sludge	reuse guidelines[1]
As	0.30 (0.11)	0.75 (0.03)	0.65 (0.09)	0.57 (0.23)	0.88 (0.06)	15
Cd	0.17 (0.08)	0.76 (0.06)	0.29 (0.14)	0.30 (0.11)	0.70 (0.05)	1.3
Cr	77 (82)	39 (39)	57 (52)	35 (24)	62 (6.0)	75
Cu	95 (51)	220 (23)	172 (68)	161 (62)	331 (23)	75
Hg	0.01 (0.004)	0.006 (0.003)	0.004 (0.000)	0.004 (0.000)	0.01 (0.003)	0.75
Ni	46 (47)	25 (24)	34 (30)	20 (13)	33 (4)	30
Pb	1.4 (0.7)	4.4 (0.7)	17 (6.0)	15 (7.0)	12 (2.6)	100
Zn	284 (85)	821 (109)	441 (133)	431 (155)	1132 (68)	300

(1) [17]

( ) standard deviation

4.3.4 Outlook

Anaerobic treatment of grey water sludge with black water increased the heavy metal content of the UASB reactor excess sludge, and might therefore hinder its soil application. In the case where soil application of the sludge outweighs the benefits of increased energy production, another treatment system for grey water should be chosen. A sequencing batch reactor (SBR) studied by Hernández Leal et al. (2010) [75] for grey water treatment produces stabilized sludge with a lower volume due to a longer SRT compared to a bioflocculation unit, and therefore benefits the disposal of the sludge. Furthermore, if grey water reuse is desired, SBR is the most favorable option due to the high effluent quality [183]. For minimum energy consumption, an anaerobic step, such as the UASB reactor, followed by an aerobic step, such as a constructed wetland (CW) or a SBR, is recommended for grey water treatment [1]. Ultimately, the origin of heavy metals in grey water should be assessed to investigate the possibility to reduce the heavy metal content, and to optimize the resource recovery from grey water.

4.4 Conclusions

Grey water sludge treatment with black water increased the energy recovery by 23% in the UASB reactor compared to black water treatment. The increase in the energy recovery can cover the increased heat demand of the UASB reactor and the electricity demand of the grey water bioflocculation system with a surplus of 0.7 kWh/cap/y electricity and 14 MJ/cap/y heat. However, grey water sludge introduced more heavy metals in the excess sludge of the UASB reactor, and might therefore hinder its soil application.

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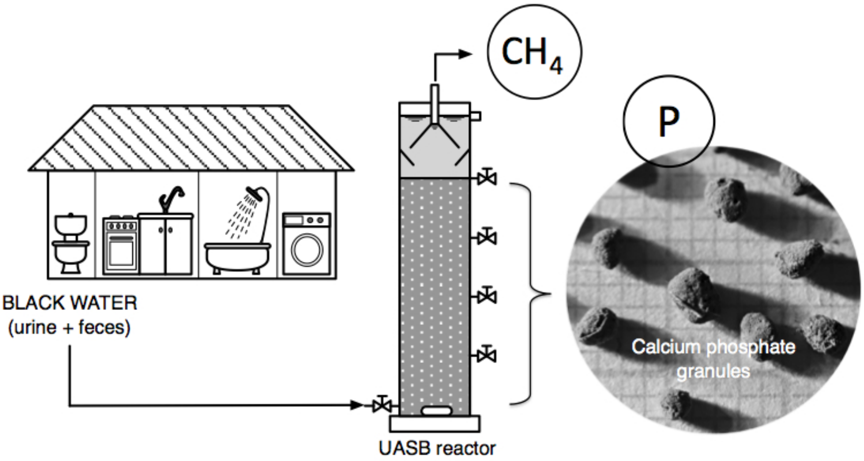
Chapter 5

Calcium phosphate granulation in anaerobic treatment of black water



Abstract

Recovery of phosphorus from wastewater as calcium phosphate could diminish the need for mining of scarce phosphate rock resources. This study introduces a novel approach to phosphorus recovery by precipitation of calcium phosphate granules in anaerobic treatment of black water. The granules formed in the upflow anaerobic sludge blanket (UASB) reactor at lab– and demonstration–scale were analyzed for chemical composition and mineralogy by Inductively Coupled Plasma–Atomic Emission Spectroscopy (ICP–AES), Electron microprobe (EMP), Fourier transform infrared spectroscopy (FTIR), Raman spectroscopy and micro X–ray Diffraction (XRD). The granules had a diameter of 1–2 mm, organic content of 33 wt%, and phosphorus content of 11–13 wt%. Three calcium phosphate phases were identified in the granules: hydroxyapatite, calcium phosphate hydrate and carbonated hydroxyapatite. Without any addition of chemicals, 7 gP/person/year can be recovered with the calcium phosphate granules, representing 2% of the incoming phosphorus in the UASB reactor. As the heavy metal content was lower compared to other phosphorus recovery products, phosphate rock and phosphorus fertilizer, the calcium phosphate granules could be considered as a new phosphorus product.

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5.1 Introduction

The production of artificial phosphorus fertilizers from phosphate rock is energy intensive and involves significant emissions of carbon, radioactive by-products and heavy metals [35]. As an alternative for mining of phosphate rock, phosphorus can be recovered from wastewater by precipitation as magnesium ammonium phosphate (struvite) $MgNH_4PO_4$ and calcium phosphate $Ca_x(PO_4)_y$ [51]. Several studies have investigated the recovery of struvite [50], its use as a fertilizer [61], and commercializing its use as a fertilizer [192]. However, the presence of ammonium, magnesium and poorly water soluble phosphate in struvite limits its use as a raw material in the phosphorus industry [51]. Instead, ongoing efforts are made to create new fertilizer markets for struvite [138]. Recovery of calcium phosphate is often considered more attractive as it has the effective composition of phosphate rock, and can therefore be processed in the phosphorus industry using the existing infrastructure.

Calcium phosphate is recovered from municipal wastewater and animal manure either by using the incinerated sludge ash where phosphorus is concentrated, or by precipitating calcium phosphate pellets in a Fluidized Bed Reactor (FBR) fed with a concentrated stream such as flushed manure [70] or a side stream of the sludge treatment [159]. The incinerated sludge ash has high heavy metal concentrations, in particular copper, zinc and iron, and cannot be processed together with phosphate rock. The calcium phosphate pellets from a FBR have lower levels of impurities and can be used by the phosphorus industry. The drawback of this technology, however, is the high consumption of chemicals to increase the solution supersaturation and to adjust the pH.

In order to minimize the operation costs, phosphorus recovery technologies should be integrated into the existing wastewater treatment processes in a way that no or a minimum amount of chemicals is needed. For example, phosphorus could be recovered in the activated sludge process without chemical additions through accumulation of hydroxyapatite (HAp) ($Ca_{10}(PO_4)_6(OH)_2$) by Polyphosphate Accumulating Organisms (PAO) in an Enhanced Biological Phosphorus Removal (EBPR) process [119]. To avoid the need for further processing of HAp into a water soluble form, calcium phosphate could be recovered in a more soluble hydrated form or as amorphous calcium phosphate (ACP) ($Ca_x(PO_4)_y{*}nH_2O$) [48]. Recovered calcium phosphate hydrate could find a niche market as a fertilizer product when the use of processed fertilizers is prohibited, such as in organic farming, provided that the level of heavy metals can be controlled.

Source separated domestic wastewater streams, such as vacuum collected black water, are ideal for phosphorus recovery due to high phosphate concentrations and relatively low heavy metal concentrations compared to municipal wastewater with industrial effluents and manure [220]. In the study of de Graaff et al. [42], phosphorus was recovered by precipitating struvite from the effluent of an upflow anaerobic sludge blanket (UASB) reactor operated on vacuum collected black water. No studies, however, have investigated the recovery of calcium phosphate within the anaerobic treatment of black water. Although de Graaff [41] reported supersaturation of HAp in black water and retainment of 39% of the incoming phosphorus in the sludge bed of the UASB reactor, no precipitation of calcium phosphate granules was observed.

This study introduces a novel approach to phosphorus recovery by precipitation of calcium phosphate granules in anaerobic treatment of black water. The chemical composition and mineralogy of the produced granules from both lab- and demonstration-scale UASB reactors are investigated by quantitative elemental analysis and direct spectral analyses. The amount of phosphorus recovered with these granules is quantified and the simultaneous phosphorus and energy recovery in the black water UASB reactor is discussed.

5.2 Materials and Methods

5.2.1 UASB reactor and black water collection

A 50 L lab-scale UASB reactor was operated for 988 days on vacuum collected black water at a hydraulic retention time (HRT) of 8.8 days and at 25°C (Figure 5.1). The reactor was started in 2010 using the same reactor configuration and operational conditions as in the study of de Graaff et al. [42], and 20 L of anaerobic sludge as an inoculum from the same study. Black water was collected with jerry cans every two weeks from 32 houses in the Decentralized Sanitation and Reuse (DESAR) demonstration site in Sneek, the Netherlands [225], and was stored at 4°C before feeding to the reactor. A demonstration-scale UASB reactor (2.4 m^3) was operated on the same black water at an HRT of 3.6 days at 35°C from 2009 on at the DESAR demonstration site [94].

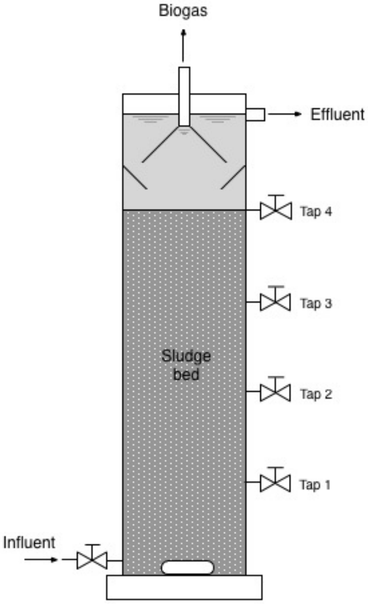

Figure 5.1: *UASB reactor configuration*

5.2.2 Influent, effluent and sludge bed sampling and analyses

Influent and effluent samples (0.5–1 L) were collected from the lab-scale UASB reactor once a week (74 samples) and analyzed immediately after collection. Total phosphorus (TP), total ammonia nitrogen (NH_4–N), anions ($PO_4{}^{3-}$, Cl^-, $SO_4{}^{2-}$, $NO_3{}^-$ and $HCO_3{}^-$), cations (Ca^{2+}, Mg^{2+}, K^+ and Na^+), total soluble phosphorus ($TP_{soluble}$), total suspended solids (TSS) and volatile suspended solids (VSS) were analyzed from the influent and effluent samples according to de Graaff et al. [42]. Total calcium (Ca_{total}) and magnesium (Mg_{total}) were analyzed from 3 influent samples using Ethos 1 Advanced Microwave digestion system of Milestone. Prior to the analysis, the influent samples were dried at 105°C overnight and the dried sample (0.4 g) was then transferred into a special microwave vessel with 10 mL of nitric acid (68 %). The samples were heated in the microwave at 180°C for 25 minutes. The acid digestion was done in duplicate. The digested samples were analyzed with Inductively Coupled Plasma-Atomic Emission Spectroscopy (ICP-AES) for calcium and magnesium. Sludge bed samples (0.25–0.5 L) taken from tap 1–4 (Figure 5.1)

(14 samples each tap) were analyzed for TSS and VSS. Additionally, 3 samples from each tap were analyzed for TP using the acid digestion and ICP-AES as described above and for $TP_{soluble}$. Excess sludge (1–10 L, 26 samples) was taken from tap 4 when the sludge bed exceeded 75 % of the reactor volume, and was analyzed for TSS and VSS, and 3 samples for TP and $TP_{soluble}$. Particulate phosphorus ($P_{particulate}$) was calculated as the difference between TP and $TP_{soluble}$. Biogas (16 samples) was analyzed according to de Graaff et al. [42].

5.2.3 Sampling and analysis of granules

The granule formation was first observed on the bottom of the lab-scale UASB reactor sludge bed after 500 days of operation, and after 750 days in the whole sludge bed. Sludge samples from the lab-scale reactor used for the following granule analyses were taken from tap 1 (Figure 5.1) at day 780 and 909. Samples from the demo-scale UASB reactor were taken only once from tap 1 after 3 years of operation due to an accidental loss of the reactor sludge bed. The granules were sieved from the sludge sample (0.5–1 L) with a 315 μm sieve, rinsed with milliQ water and air dried overnight at room temperature to prevent dehydration of the crystal structure. To determine the organic content of the granules, 0.2 g of dried granules (105°C) (done in triplicate) from the lab-scale reactor were burned at 550°C and the difference in the weight was recorded.

Quantitative elemental analysis was done on the granules from both lab- and demo-scale reactors combining acid destruction and ICP-AES. The dried granule samples (0.5 g per sample) were acid destructed in 10 mL of nitric and hydrochloric acid (mixture ratio 1:3) using the microwave digestion system described above. The acid destruction was done in duplicate only with the lab-scale samples as there was not enough sample material from the demo-scale reactor. These samples were then analyzed with ICP-AES for calcium, phosphorus, magnesium, potassium, iron and heavy metals (zinc, copper, arsenic, cadmium, chromium, mercury, nickel and lead).

Electron microprobe (EMP) analysis was used to determine the spatial distribution of elements in thin sliced sections of 3 dried granules from both lab- and demo-scale reactors. Prior to the analysis the thin sliced sections were polished and mounted in epoxy. The EMP analysis was performed with a JEOL JXA 8800M microprobe operated at 20kV accelerating voltage and 20 nA probe current using a static focused beam. The quantitative data was obtained from 1 granule (lab-scale) using standards from Jarosewich [81] for the calibration.

Fourier transform infrared spectroscopy (FTIR), Raman spectroscopy and micro X-ray Diffraction (XRD) was used to analyze precipitated species in the granules from both lab- and demo-scale reactors. The FTIR and Raman analysis was done on 3 dried granules per reactor, and the XRD analysis was done on 2 dried granules from the lab-scale and 1 dried granule from the demo-scale reactor. The Raman analysis was also done on 1 burned granule (550°C) from both lab- and demo-scale reactors, from which calcium carbonate clusters were identified. The granules were slightly crushed to reveal the inner part prior to the analysis. The FTIR analysis was performed using Shimadzu 8400S and the Raman analysis was performed with a high resolution LabRAM from Horiba Jobin Yvon using a 532 nm laser beam and an Olympus microscope (objective of 50 times). Reference spectra used in the analyses were taken from the RRUFF library. The XRD analysis was performed using Bruker D8 operated at 35 kV × 45 mA with a beam diameter of 300 μm.

5.2.4 Phosphorus mass balance calculations

The phosphorus mass balance of the lab-scale UASB reactor was established over the whole operation period of 988 days according to Eq. (5.1)

$$P_{influent} = P_{effluent} + P_{sludge\ bed} + P_{wasted\ sludge} \tag{5.1}$$

where $\mathrm{P}_{influent}$ and $\mathrm{P}_{effluent}$ are the accumulative phosphorus load in influent (black water) and effluent, respectively (in gP), $\mathrm{P}_{sludge\ bed}$ is the amount of phosphorus accumulated in the sludge bed (in gP) and $\mathrm{P}_{wasted\ sludge}$ is the amount of phosphorus wasted with excess sludge and sludge sampling (in gP).

The amount of phosphorus in the granules in the sludge bed and wasted sludge was calculated according to Eq. (5.2)

$$P_{granules}(g) = TSS_{granules}(g/L_{sludge}) * V_{sludge}(L) * P_{granules(ICP-AES)}(\%) \tag{5.2}$$

where TSS is the solids concentration of the granule samples taken from tap 1–4 (sieved with a 315 μm sieve from the sludge sample), V_{sludge} is the volume of the sludge bed (75 % of 50 L reactor) and wasted sludge (including only the time period when granules were observed), and $\mathrm{P}_{granules(ICP-AES)}$ is the phosphorus content of the granules obtained from the quantitative elemental analysis (ICP-AES).

The amount of phosphorus incorporated in the biomass in sludge bed and wasted sludge was calculated with a typical phosphorus content of bacteria cells of 20 mgP/g dry weight [181].

The total amount of phosphorus recovered with the granules (gP/person/year) was determined using a UASB reactor capacity of 0.95 person according to the average black water loading of the reactor of 5.7 L/d and the black water production of 6 L/person/d [225].

5.2.5 Saturation index calculations

Saturation Index (SI) calculations for calcite (as an indicator for calcium carbonate precipitation), HAp (as an indicator for calcium phosphate precipitation) and struvite in tap water and black water were performed with the OLI Stream Analyzer (version 3.1, [137]). Saturation index was defined according to Eq. (5.3)

$$SI = \frac{IAP}{K_{sp}} \tag{5.3}$$

where IAP is the ion activity product and K_{sp} is the solubility product of the precipitate phase. The solution is considered to be unsaturated when the SI is below zero, at saturation when zero, and supersaturated when the SI is above zero.

5.3 Results

5.3.1 Granule structure and elemental composition (ICP-AES and EMP)

Granule formation was observed in the flocculent sludge bed of the lab-scale black water UASB reactor after 500 days of operation (Figure 5.2 a). The dried granules have a diameter of 1–2 mm (Figure 5.2 b) and a white inorganic core of a similar diameter after burning away the organic content at 550°C (Figure 5.2 c). The average organic content of the granules is 33 wt% (s.d. 1.5) and is located mainly in the outer layer of the granules. Same type of granules are also found in the demo-scale UASB reactor at the DESAR demonstration site. The exact onset time of the granule formation in the demo-scale reactor is not known.

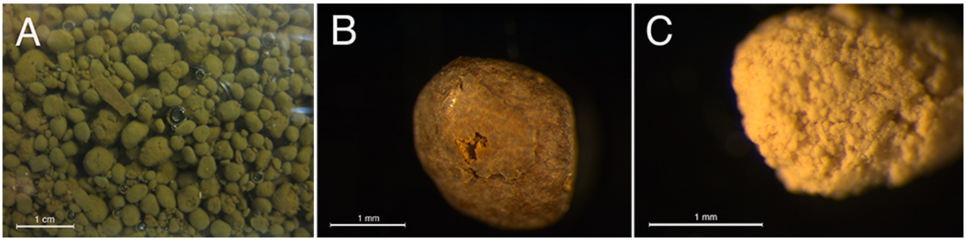

Figure 5.2: *Granule formation in the black water UASB reactor (a) dried granule (b) and inorganic core of a granule (burned at 550° C) (c)*

Table 5.1 presents the quantitatively analyzed elemental composition of the whole granules (ICP-AES) from both lab- and demo-scale reactors, and of different areas within a thin sliced section of a granule (EMP) from the lab-scale reactor. The relative standard deviation (%RSD) of the duplicates in the acid destruction and ICP-AES analysis is within 5 %. The prominent elements in the whole granules are calcium (21–22 wt%) and phosphorus (11–13 wt%), with traces of magnesium (0.3 wt%) and potassium (0.05–0.06 wt%). The molar Ca/P ratio of the whole granules from the lab-scale (1.58) is similar to the theoretical ratio of ACP (1.5) [32], while the ratio of the granules from the demo-scale (1.25) is similar to the theoretical ratio of octacalcium phosphate (OCP) (1.3) ($Ca_8(HPO_4)_2(PO_4)_4$*$5H_2O$) that is an intermediate phase in the development of HAp [48]. Precipitation of struvite or potassium ammonium phosphate in the granules is not expected due to the low measured magnesium and potassium concentrations.

Table 5.1: *Quantitative elemental composition of granules (ICP-AES and EMP)*

Element	Unit	ICP-AES whole granule (lab-scale)	 (demo-scale)	EMP thin sliced section (lab-scale) area (A)	 area (B)
Ca	wt%	22	21	11	11
P	wt%	11	13	4.7	4.7
Mg	wt%	0.3	0.3	0.3	0.4
K	wt%	0.05	0.06	0.01	0.02
molar Ca/P ratio		1.58	1.25	1.81	1.81

The quantitatively analyzed areas within the thin sliced section of the granule are presented in Figure 5.3. The area (A) is specified as the general matrix of the inner part of the granule with prominent elements being calcium (11 wt%) and phosphorus (4.7 wt%), and with traces of magnesium (0.3 wt%) and potassium (0.01 wt%). The molar Ca/P ratio of 1.81 is higher compared to the ratios attained with the ICP-AES analysis and to the theoretical ratio of HAp (1.67), and could originate from local small calcium carbonate ($CaCO_3$) clusters within the calcium phosphate matrix. Another explanation for the higher Ca/P ratio could be presence of calcium deficient carbonated HAp ($Ca_{10}(PO_4)_3(CO_3)_3(OH)_2$) with a theoretical Ca/P ratio of 3.33. The area (B) is specified as part of a biopolymer spiral structure inside the granule with calcium phosphate and calcium carbonate clusters attached to it (molar Ca/P ratio of 1.81). Other elements, such as sulfur, sodium, silica, aluminum, manganese, iron and heavy metals were below 1 wt% in both ICP-AES and EMP analysis. However, randomly distributed micro-clusters of sodium, silica, aluminum, iron and heavy metals, such as copper, zinc, chromium and nickel, were detected in the thin sliced sections of the granules with the EMP analysis. Heavy metals and iron are further quantified under the sub-chapter 5.3.4.

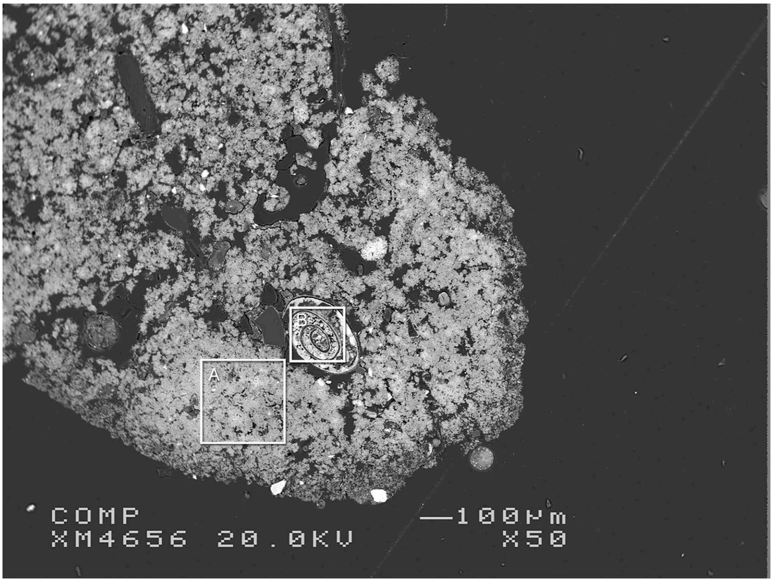

Figure 5.3: *Thin sliced section of a granule from the lab-scale reactor with quantitatively analyzed areas*

Figure 5.4 demonstrates the distribution of calcium and phosphorus over the thin sliced sections of granules from both lab- and demo-scale reactors. Evenly distributed calcium and phosphorus over the granule sections suggest a homogenous calcium phosphate matrix. Separate small clusters of calcium carbonate within the calcium phosphate matrix are determined by high local calcium levels and absence of phosphorus (red indicates high and blue indicates low concentration). The predominance of calcium carbonate over dolomite ($CaMg(CO_3)_2$) is verified by absence of magnesium in these clusters of high calcium levels (images not shown). Similar calcium carbonate clusters within the calcium phosphate matrix are also identified in Raman analysis (see sub-chapter 5.4.2). The elemental mapping images further reveal an edge with low levels of calcium and phosphorus, confirming an organic outer layer that contributes to the organic content of the granules.

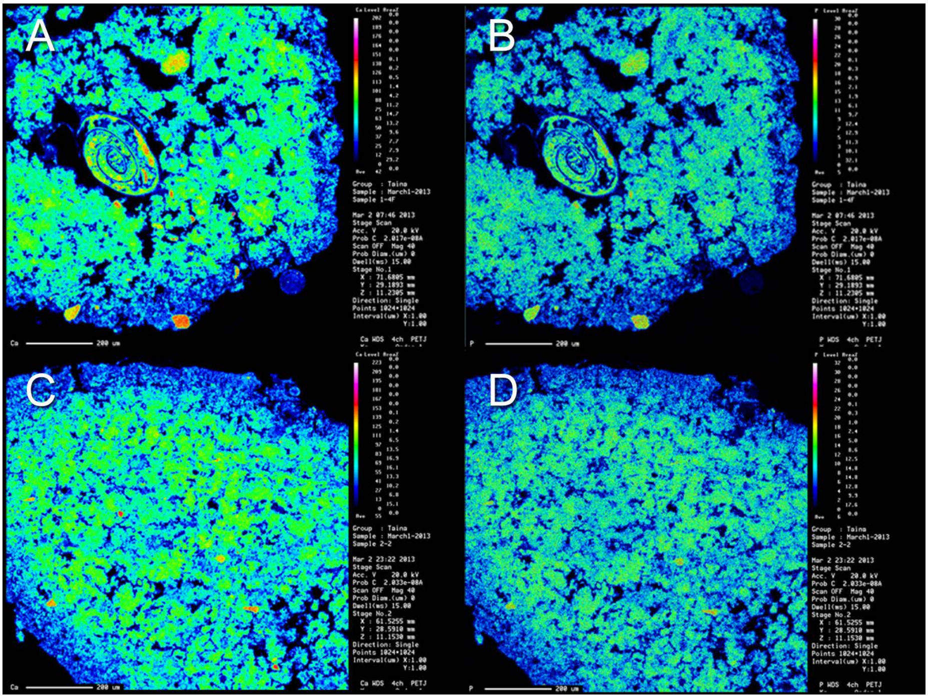

Figure 5.4: *Elemental mapping of calcium (a & c), and phosphorus (b & d) in thin sliced sections of the granules from lab- (a & b) and demo-scale (c & d) reactors*

5.3.2 Precipitated species in granules (FTIR, Raman and XRD)

Figure 5.5 presents the XRD pattern of one of the unheated granules from the lab-scale reactor where three possible calcium phosphate phases are distinguished; HAp, calcium phosphate hydrate ($Ca_3(PO_4)_2{*}H_2O$) and carbonated HAp. The same phases are identified in the granules from both lab- and demo-scale reactors. These calcium phosphate phases are in accordance with the ICP-AES data suggesting presence of ACP or calcium phosphate hydrate with the same molar Ca/P ratio (1.5) and OCP that readily transforms into HAp, and with the EMP data suggesting presence of carbonated HAp that contributes to the higher molar Ca/P ratios.

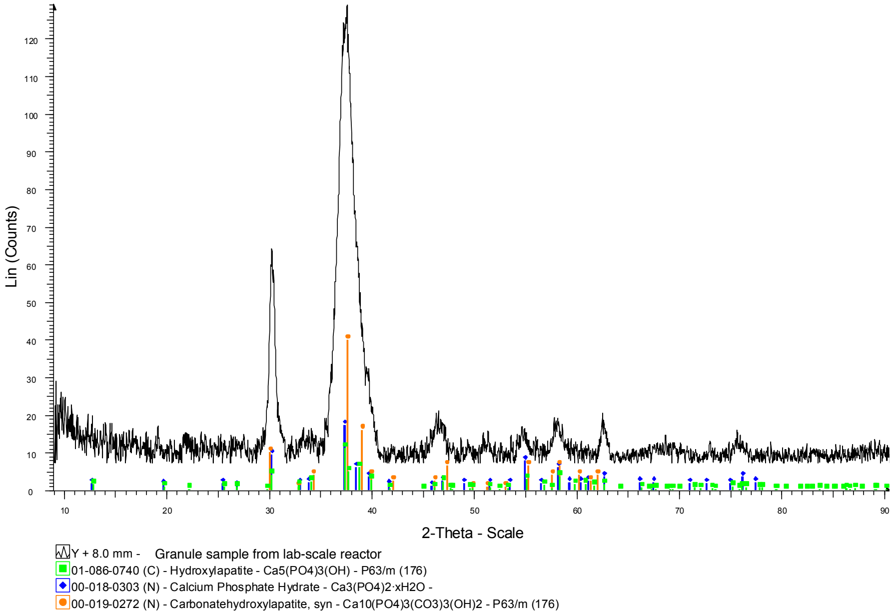

Figure 5.5: *XRD pattern of a granule from the lab-scale reactor*

Figure 5.6 presents the Raman spectra of the unheated granules from both lab- and demo-scale reactors and the reference spectra of HAp. Similar to the XRD pattern, the Raman spectra indicates presence of calcium phosphate hydrate in the granules with the PO_4 v_1 band located at around 950 cm^{-1}, 10 cm^{-1} lower than that of HAp

[32]. The PO_4 v_1 band varies in the range 945–948 cm^{-1} with the granules from the lab-scale reactor and in the range 947–958 cm^{-1} with the demo-scale reactor, the highest band being close to the one of HAp (960 cm^{-1}). The higher temperature in the demo-scale reactor (35°C) might enhance the transformation of calcium phosphate hydrate into more stable HAp [19]. The reactor temperature therefore appears to be an important parameter defining the crystallinity of the produced calcium phosphate granules.

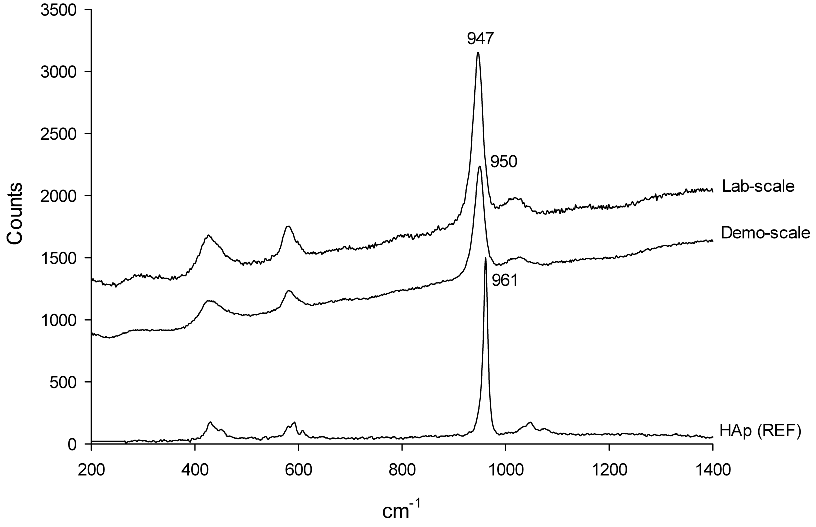

Figure 5.6: *Raman spectra of the granules from lab- and demo-scale reactors and the reference spectra of HAp*

Figure 5.7 presents the FTIR spectra of the unheated granules from both lab- and demo-scale reactors and the reference spectra of HAp. In the spectra of the granules the PO_4 v_1 band is located at 960 cm^{-1} and the PO_4 v_3 band at 1010 cm^{-1}, distinctive to the spectra of HAp. However, the broad bands at around 3300 cm^{-1} refer to presence of H_2O in the crystal structure [4], and thus to a poorly crystalline calcium phosphate, such as calcium phosphate hydrate, also identified in the XRD pattern and the Raman spectra. The presence of carbonate in the granules can be related to the peaks at 870 cm^{-1} [4] and 1300–1650 cm^{-1} [148]. The two peaks at 1230 cm^{-1} and 1260 cm^{-1} are distinctive to zinc, copper and organometallics, and can be explained by the presence of organics in the outer layer of the granules and wide variety of metals detected in the ICP-AES and EMP analysis.

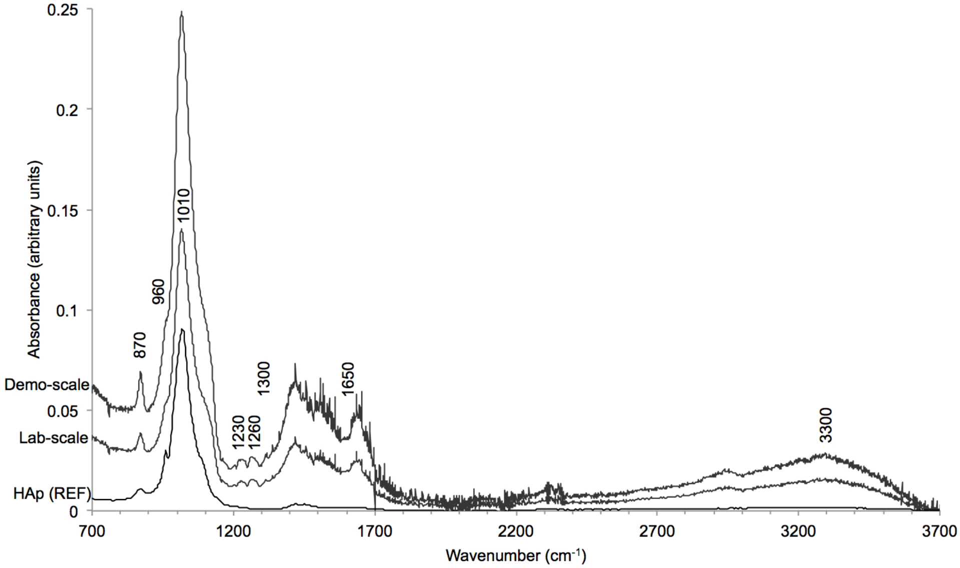

Figure 5.7: *FTIR spectra of the granules from lab- and demo-scale reactors and the reference spectra of HAp*

5.3.3 Phosphorus mass balance

The phosphorus mass balance of the UASB reactor is determined over the whole operation period of 988 days. From the incoming phosphorus (1006 gP from which 2 % is inoculum) 49 % (489 gP) remains in the effluent, 20 % (200 gP) is wasted as excess sludge and sludge sampling, and 6 % (60 gP) is retained in the sludge bed. The granules represent 16 % (9.6 gP) of the phosphorus retained in the sludge bed and 5 % (9.4 gP) of the phosphorus in the wasted sludge. According to the phosphorus mass balance, the amount of phosphorus recovered with the granules is 7 gP/person/year, representing 2 % of the incoming phosphorus. However, the mass balance is not fully closed as 26 % (258 gP) of the incoming phosphorus is missing. The possible sources for the missing phosphorus are large amount of phosphate precipitate on top of the reactor in the solid-liquid-gas separator, and settling of the granules on the bottom of the reactor below the first sludge bed sampling point (Tap 1 in Figure 5.1).

Figure 5.8 presents the fractionation of particulate phosphorus between the granules, biomass and inorganic particles ($\leq$315 μm) in the sludge bed. The largest fraction of phosphorus in the granules is in the bottom part of the sludge bed (Tap 1) and the smallest fraction is in the top part of the sludge bed (Tap 4), indicating settling of the granules on the bottom of the reactor, and could therefore hold a large portion of the missing phosphorus. The fraction of particulate phosphorus not included in the granules and biomass is considered to consist of inorganic particles ($\leq$315 μm), and this fraction accounts for 23 % of the particulate phosphorus in the sludge bed. By accounting these small inorganic particles as the initial state of the granules, the total amount of phosphorus recovered with the granules would increase to 12 gP/person/year, representing 3 % of the incoming phosphorus.

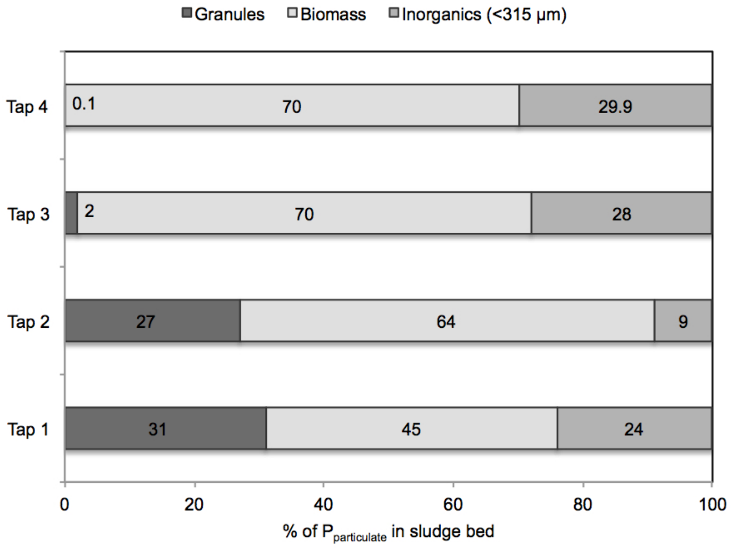

Figure 5.8: *Fractionation of particulate phosphorus between the granules, biomass and inorganic particles ($\leq$315 μm) in the sludge bed of the UASB reactor*

5.3.4 Calcium phosphate granules as candidate phosphorus product

Table 5.2 presents the phosphate, iron and heavy metal content of the calcium phosphate granules, other phosphorus recovery products, phosphate rock, a common phosphorus fertilizer Triple Superphosphate (TSP), and the requirements for phosphorus industry. The %RSD of the duplicates in the acid destruction and ICP-AES analysis is within 5 %. The calcium phosphate granules have a comparable or higher phosphate content compared to the other phosphorus recovery products, phosphate rock and TSP. The granules from both lab- and demo-scale reactors and the Crystalactor pellets are in accordance with the requirements for phosphorus industry, leaving out the sewage sludge ash with too high iron, zinc and copper concentrations. Due to the source-separation of black water from the sewage and industrial effluents, the granules from both reactors have lower concentrations of iron, zinc, arsenic and cadmium than the Crystalactor pellets originating from a side stream of the sewage sludge treatment. However, the copper concentration in the granules is higher than in other products and the origin of copper in black water needs to be further determined. Nevertheless, the calcium phosphate granules have lower concentrations of zinc, arsenic, cadmium, chromium and nickel, and comparable concentrations of mercury and lead to phosphate rock and TSP, and could be considered as a new phosphorus product. In addition, unlike the use of artificial fertilizers, no radioactive by-products are involved in the use of calcium phosphate granules. However, as the organic content of the raw material is one of the limiting parameters for phosphorus industry [51], a pre-treatment step to remove the organic outer layer of the granules might be needed. Further investigations should also be done on the bioavailability of the granules to evaluate their value as a fertilizer product.

Table 5.2: *Phosphate, iron and heavy metal content of calcium phosphate granules, other phosphorus recovery products, phosphate rock, Triple Superphosphate, and requirements for phosphorus industry*

Parameter	Unit	P granules (lab-scale)	P granules (demo-scale)	P pellets Crystalactor^1	Sewage sludge ash^2	P industry^2	P rock^3	TSP^3
P_2O_5	wt%	35	39	26	36	$\geq$25	40	46
Fe	g/kgDW-P_2O_5	2	2	5	44	$\leq$40	-	-
Zn	mg/kgDW-P_2O_5	430	812	1192	8611	$\leq$4000	1120	770
Cu		143	182	65	4167	$\leq$2000	80	33
As		4.3	4	8	-	-	25	13
Cd		4.7	3	23	-	-	64	62
Cr		32	93	31	-	-	380	498
Hg		0.06	0.05	-	-	-	0.08	0.04
Ni		29	56	31	-	-	93	46
Pb		23	17	-	-	-	10	33

(1) Driver et al. [51]
(2) Schipper et al. [159]
(3) Dittrich and Klose [46]
- not determined

5.4 Discussion

5.4.1 Chemical composition and mineralogy of granules

According to quantitative analysis, the main elements in the granules are calcium and phosphorus. Direct spectral analyses present three different calcium phosphate phases precipitating in the granules: HAp, calcium phosphate hydrate and carbonated HAp. Within the calcium phosphate matrix, several small calcium carbonate clusters are revealed by elemental mapping and Raman analysis. To retain the purity of calcium phosphate, a minimum amount of carbonate is desired in the granules. The low calculated carbonate content of 1.3–4.9 wt% in the granules is based on stoichiometry of two scenarios: carbonate originates solely from the calcium carbonate clusters within ACP/HAp matrix, or from the carbonated HAp. The exact carbonate content of the granules need to be further experimentally determined.

5.4.2 Initiation of calcium phosphate precipitation in black water UASB reactor

Supersaturation of HAp in black water was recorded in the study of de Graaff [41], but no formation of calcium phosphate granules was observed using a UASB reactor operated for a period of 951 days under exactly the same conditions as in the present study. Following hypotheses on the combined effect on the initiation of calcium phosphate precipitation in the UASB reactor are formulated:

1. Changes in the chemical composition of tap water used for toilet flushing and black water increase the supersaturation state of mineral species and lead to formation of seed crystals
2. Biofilm surrounding the granules in the UASB reactor creates a micro-environment that controls the diffusion of ions and elevates the pH locally, inducing the precipitation of calcium phosphate in the granules

Table 5.3 presents the tap water and black water compositions in this study and in the study of de Graaff et al. [42] used for the SI calculations, and the SI of HAp, calcite and struvite calculated with the OLI Stream Analyzer. The increased bicarbonate and free calcium concentrations in tap water in 2011 compared to 2010 slightly increase the SI for calcite, and could have led to formation of calcium carbonate seed crystals. This assumption is supported by the elemental mapping and Raman analysis of the granules that reveal several small clusters of calcium carbonate within the calcium phosphate matrix (Figure 5.9 and 5.10). The presence of seed crystals catalyze the precipitation of calcium phosphate by reducing the energy barrier for the nucleation process [169]. In black water the decreased bicarbonate concentration and the increased free calcium concentration in this study compared to the study of de Graaff et al. [42] create favorable conditions for precipitation of calcium phosphate. Although the SI of calcite and struvite also increases slightly, the significantly higher SI of HAp indicates the predominance of calcium phosphate precipitation in black water. Furthermore, phosphate at soluble inorganic phosphorus concentration $\geq$1.5 mg/L (49 mg/L in this study) is known to obstruct the growth of calcite and to form a separate calcium phosphate phase [145]. These changes in tap water and black water coincide with the installation of an ion exchanger as an additional treatment step to remove soil organic acids from tap water in the drinking water treatment facility supplying water to the toilets producing black water in the DESAR demonstration site in 2010. Soil organic acids, such as humic and fulvic acids, inhibit the precipitation of mineral species in two ways; decreasing the solution supersaturation by binding free ions and decreasing the precipitation kinetics by adsorbing onto the crystal growth sites [4, 110]. The changes in the tap water and black water compositions seem to therefore enhance the precipitation of calcium phosphate not only thermodynamically, but also kinetically.

Table 5.3: *Tap water and black water compositions, and SI of HAp, calcite and struvite*

Parameter	Unit	Tap water 2010[1]	Tap water 2011[1]	Black water[2]	s.d.	Black water (This study)	s.d.
T	°C	11.5	11.5	25		25	
pH		8.25	8.23	8.7		8.1	
HCO_3^-	mg/L	281	292	5612	2791	3628	1129
NH_4^+	mg/L	nd	nd	1414	193	1157	154
PO_4^{3-}	mg/L	nd	nd	204	54	149	28
Ca^{2+}	mg/L	31	38	39	18	62	7.4
Mg^{2+}	mg/L	11	10	4	4.9	15	6.7
K^+	mg/L	2	2	424	87	287	30
Na^+	mg/L	84	79	604	134	426	34
Cl^-	mg/L	29	30	695	138	506	95
SO_4^{2-}	mg/L	2	2	157	42	55	35
NO_3^-	mg/L	13	12	26	24	0.7	1.5
SI							
HAp		-	-	$2.1*10^8$		$1.9*10^9$	
Calcite		0.54	0.63	17		21	
Struvite		-	-	0.4		1.2	

(1) Vitens [210]
(2) de Graaff et al. [42]
s.d. = standard deviation
nd = not determined
- undersaturation

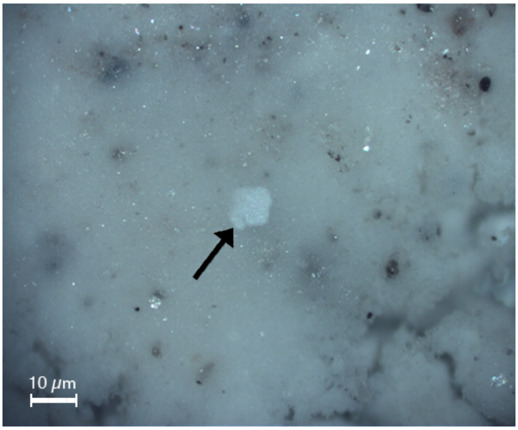

Figure 5.9: *Raman picture of a calcium carbonate cluster within the calcium phosphate matrix of a lab-scale granule*

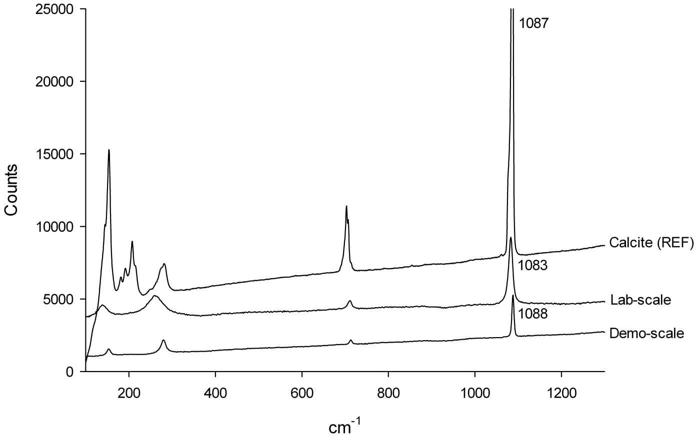

Figure 5.10: *Raman spectra of the calcium carbonate clusters and the reference spectra of calcite*

Carbonate and magnesium, abundantly present in black water, are known to inhibit the precipitation of calcium phosphate by competing for the active growth sites on the crystal lattice with phosphate and calcium ions, respectively [120]. However, calcium phosphate is found to be the dominant species in the granules with low levels of carbonate (1.3–4.9 wt%) (calculated) and magnesium (0.3–0.4 wt%). Although the solubility product of calcium phosphate ($1.0*10^{-36}$) is significantly lower compared to calcite ($3.8*10^{-9}$) and struvite ($2.5*10^{-13}$), the precipitation of calcite and struvite could be kinetically more favorable. An explanation for the predominance of calcium phosphate could be the biofilm around the granules (Figure 5.2 and 5.4) creating a micro-environment in which the diffusion of ions is controlled and a local pH gradient is formed due to conversion of acetate (pK_a 4.76) into carbonate (pK_a 6.37). Beside enriching phosphate at elevated pH, the long retention times provided by the biofilm around the granules could support the growth of calcium phosphate precipitate into mature granules. In addition, regardless of the inhibitory nature of carbonate and magnesium, the coexistence of these ions eliminates the individual inhibitory effect due to formation of magnesium carbonate ($MgCO_3$), and thus reduction in free carbonate and magnesium activities [26]. The formation of magnesium carbonate in this study could be related to the decrease in pH and bicarbonate concentration, and the increase in the solid fraction of magnesium in black water. In the ongoing research the biotic and a-biotic factors affecting calcium phosphate granule formation in the black water UASB reactor are investigated.

5.4.3 Simultaneous phosphorus and energy recovery from black water

Without any addition of chemicals, phosphorus can be recovered 7 gP/person/year as calcium phosphate granules from the black water UASB reactor. As it represents only 2 % of the incoming phosphorus, additional recovery of phosphorus from the effluent is recommended to improve the recovery ratio and effluent quality. For the recovery of calcium phosphate granules to be a realistic alternative for the struvite recovery, the possible optimization of this process by addition of calcium is investigated in the ongoing research.

Simultaneously with phosphorus recovery, methane is recovered 1.7 m^3 CH_4/m^3 black water, comparable to the amount of methane recovered in the study of de Graaff et al. [42] (1.8 m^3 CH_4/m^3 black water). As the biomass concentration in the reactor increases from 19 to 29 gVSS/$L_{reactor}$, a higher loading can be applied to further save energy in the reactor configuration. The increased biomass retention can be explained by attachment of microbial aggregates on an inert carrier material, such as mineral particles, and further formation of dense granular sludge [79].

The present study shows that phosphorus and energy can be simultaneously recovered in the anaerobic treatment of black water. Further research is required to determine a practical application to separate the calcium phosphate granules from the reactor sludge bed without interfering the digestion process of black water.

5.5 Conclusions

For the first time, calcium phosphate granules were observed in the UASB reactor operated on black water at both lab-and demo-scale, and were analyzed for chemical composition and mineralogy.

- The dried granules had a diameter of 1–2 mm, organic content of 33 wt%, and phosphorus content of 11–13 wt%.
- According to direct spectral analysis the prominent precipitated species in the granules were calcium phosphate hydrate, HAp and carbonated HAp.
- Without any addition of chemicals, 7 gP/person/year was recovered with the calcium phosphate granules, representing 2% of the incoming phosphorus in the UASB reactor.
- The calcium phosphate granules had a lower heavy metal content compared to other phosphorus recovery products, phosphate rock and phosphorus fertilizer, and could be considered as a new phosphorus product.

This study advances a new approach to phosphorus recovery in which calcium phosphate is simultaneously recovered with energy from black water. In the ongoing research the biotic and a–biotic factors affecting calcium phosphate granule formation in the black water UASB reactor are investigated.

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Chapter 6

Concluding discussion

6.1 Organic fertilizer recovery

6.1.1 Closing the soil/food heavy metal cycle

In order to close the soil/food heavy metal cycle, external input of heavy metals to agriculture should be limited. As presented in Chapter 3, feces and urine are the main contributors to the heavy metal loading of vacuum collected black water, while most of the heavy metals in sewage originate from industrial effluents, surface run-offs and tap water. Since heavy metals in feces and urine primarily originate from food products, soil application of black water sludge should be practised to close the soil/food heavy metal cycle. To distinguish black water from sewage in sludge reuse regulation, a control parameter should be established, such as the Hg and Pb content that is significantly higher in sewage sludge compared to black water sludge (from 50- to 200-fold). In addition, livestock manure and artificial fertilizers should also be included in the heavy metal regulation. The level of heavy metals and micropollutants in manure is higher compared to feces and urine [220], and further efforts should be made to decrease the input of heavy metals and micropollutants to livestock feed. The Cd content is significantly lower in black water sludge compared to the average levels in phosphate fertilizers within Europe [135], and the heavy metal content of phosphate rock used for artificial fertilizer production should be therefore controlled.

Co-digestion of different biosolids and waste streams is a common practise to increase energy recovery [121]. When treating a mixture of different waste streams, attention should be also given to the quality of excess sludge in terms of heavy metals and other pollutants, to ensure soil application of the sludge. Addition of kitchen refuse to anaerobic treatment of black water improves methane production [115]. However, the elevated heavy metal content of kitchen refuse, in particular Cd, Cu, Pb and Zn [204], might deteriorate the excess sludge quality, and further hinder the soil application of black water sludge. Nevertheless, the heavy metal content of kitchen refuse is also directly linked to the soil/food cycle, and therefore influenced by the application of manure and artificial fertilizers in agriculture. From this viewpoint, the soil application of black water sludge could reduce the amount of heavy metals in the soil/food cycle, and subsequently reduce the heavy metal content of kitchen refuse.

To further improve energy recovery within source-separated sanitation concepts, the organic matter present in grey water can be utilized in an integrated treatment system combining anaerobic treatment of grey water sludge with black water. Chapter 4 demonstrated increased methane production in the UASB reactor operated on the mixture of grey water sludge and black water by introducing an additional organic fraction, and by increasing the methanization. However, grey water sludge introduced more heavy metals in the excess sludge, and might therefore hinder its soil application. To eliminate external heavy metal sources, such as household chemicals from black water, it is therefore not recommended to mix it with grey water. Closing of the organic carbon cycle should be given priority over energy recovery, also stated in the European Waste Directive [54]. Micropollutants, such as personal care and household products primarily present in grey water [73], are of emerging concern in the urban water cycle, and separate treatment of grey water sludge should be established to meet the evolving sludge reuse regulation on micropollutants [87]. To continue the work of de Graaff et al. (2011) [44], further research is also required to address the issue of micropollutants, such as pharmaceuticals and hormones in black water.

6.1.2 Black water sludge reuse – disinfection and soil application

Hygiene is one of the most important aspects of sanitation systems. Centralized sanitation systems have approached the problem via transport and dilution, yet fail to ultimately protect human health from pathogenic organisms introduced via sewage leakages, overflows and contaminated water sources [174]. The significantly lower volume of vacuum collected black water compared to sewage (50–fold as presented in Chapter 2) might benefit the disinfection of this stream. To further disinfect black water sludge after anaerobic treatment, several treatment technologies used for human excreta can be applied, such as storing, dehydration and composting [160, 209, 134]. Composting of black water sludge, however, requires an additional carbon source, and it could be implemented as co-composting with locally available garden waste. Alternatively, hyper-thermophilic treatment at 70°C can be used to pasteurize black water [114], and can be energy efficiently applied using vacuum collection with minimum amount of flushing water [224].

Important part of introducing new fertilizer products to farmers is the possibility of using existing equipment for spreading the product on agricultural soil. Similar to handling of human excreta [84], disinfected black water sludge can be spread on agricultural soil with the existing system for livestock manure and slurry, provided that the water content is low enough. Emissions originating from the use of organic fertilizers are often connected to the risk of eutrophication and acidification. However, the use of human excreta as organic fertilizer involves less greenhouse gases than the complete life cycle of artificial fertilizers [172]. Furthermore, anaerobically treated black water involves less emission compared to raw waste due to the use of stabilized sludge.

In order to attain successful reuse of black water sludge in agriculture, acceptance from both farmers and consumers are of importance. Concerns about source-separation and the use of treated toilet waste in agriculture are mostly connected to the efforts required for maintenance of the toilet, wastewater collection and transport, and the safety of the new fertilizer product [109]. Cooperation between the different stakeholder groups of building companies, agricultural sector, water boards, and the government is needed to support the integration between the social and technological aspects of source-separation and new fertilizer products [100]. More and more environmental awareness and interest towards ”self-sufficient living” is taking place in a modern, urbanized society with people yearning for connection with nature. Popular books, such as *Humanure* by Jenkins (2005) [82] on do-it-yourself composting of human excreta, and *The Big Necessity* by George (2008) [62] on a new perspective on human waste, are reflections of the new mindset on returning back to the fundamentals of life.

6.2 Phosphate recovery

6.2.1 High quality phosphate product

Recovery of struvite over calcium phosphate is often practised due to more favorable precipitation conditions, such as wider pH range and molar Mg/Ca ratio, slow nucleation kinetics resulting in growth of larger particles with improved settling characteristics, and a higher phosphate removal efficiency [132]. Opposed to this, precipitation of high purity calcium phosphate requires an Mg/Ca ratio of ≤ 0.1, and the fast nucleation kinetics result in precipitation of fines, leading to problematic separation from the supernatant. However, as presented in Chapter 5, calcium phosphate can also be recovered as granules at an Mg/Ca ratio of 0.4 and with a high purity of $\geq 95\%$, in contrast to the study of Muster et al. (2013) [132]. The occurrence of high purity calcium phosphate granules instead of fines can be attributed to the simultaneous influence of chemical, physical and biological factors in the sludge bed of the upflow anaerobic sludge blanket (UASB) reactor operated on vacuum collected black water.

Water solubility of the phosphate product, representing the bioavailability of phosphate to plants, is an important characteristic whether the product will be used directly as a fertilizer or as raw material in fertilizer industry. To turn phosphate rock into a water soluble phosphate fertilizer, such as Triple Super Phosphate (TSP), addition of phosphoric acid is required. Therefore, the higher the water solubility of the phosphate product, the less chemicals are needed to produce a fertilizer product. The calcium phosphate granules presented in Chapter 5 have a high porosity, and are partly composed of hydrated calcium phosphate, indicating high water solubility. However, further experiments are required to determine the water solubility of the granules, and compare it to other phosphate recovery products and phosphate rock.

6.2.2 Optimizing calcium phosphate granulation

The goal of optimal phosphate recovery is production of high purity product in an energy efficient reactor configuration with a minimum addition of chemicals. The formation of high purity calcium phosphate granules in the anaerobic treatment of black water is influenced by thermodynamical parameters, such as sufficient ion concentration and pH, and physical parameters, such as long retention time and possible presence of seed crystals. However, as discussed before, in purely chemical/physical processes calcium phosphate precipitates as fines, and therefore, the biological factors play a key role in the granulation process. In this process, biofilm is formed around the granules, creating a micro-environment inside the granules. According to the hypothesis, pH is increased locally in the micro-environment as a result of bacterial metabolism, creating more favorable conditions for calcium phosphate precipitation compared to supernatant in the sludge bed. Furthermore, the biofilm controls the diffusion of ions, and the decay of organic molecules containing phosphate enhance the concentration of phosphate inside the granules. The sludge concentration is increased due to granulation, increasing the sludge retention time (SRT) in the UASB reactor, and further enhancing the growth of calcium phosphate granules.

In the process presented in this thesis, phosphate is recovered in the sludge bed of the UASB reactor, and no separate treatment unit is required. In this way, both energy and phosphate can be simultaneously recovered in one anaerobic reactor. In most of the calcium phosphate recovery processes, the co-precipitation of calcium carbonate is minimized by addition of either acid or base to control the carbonate content and pH. However, when the ion concentration and pH are controlled by a biofilm, no additional chemicals are needed to guarantee the purity of produced calcium phosphate granules.

To increase the phosphate recovery rate in the UASB reactor, different chemical, physical and biological factors can be optimized. A small amount of additional calcium can be dosed to increase the supersaturation of calcium phosphate. To increase the retention time, recirculation of the granular sludge can be applied. Addition of seed material, such as calcite beads, can be used to enhance calcium phosphate precipitation by lowering the nucleation energy. These chemical and physical factors can be integrated in a fluidized bed reactor (FBR), where the granulation process is optimized due to better distribution of seed material and additional calcium [166]. This process promotes a better supersaturation profile, aggregation of particles, and separation of granules based on their size. In terms of biological factors, selective use of micro-organisms, such as acetate converting methanogens, can increase the calcium phosphate granulation by enhanced metabolic activity.

6.3 Proposed sanitation concept and future research

Based on the information gathered in this thesis, a sanitation concept is proposed for recovery of organic fertilizer, phosphate, energy and water from black and grey water (Figure 6.1). Emphasis is laid on the improved recovery of organic fertilizer and phosphate from black water.

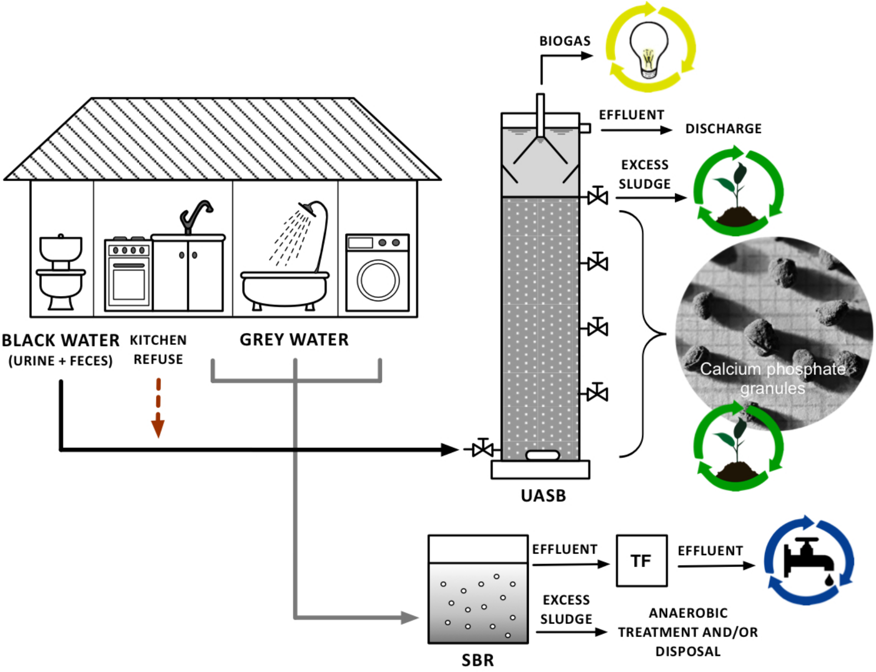

Figure 6.1: *Proposed sanitation concept with recovery of organic fertilizer, phosphate, energy and water from black and grey water (UASB = upflow anaerobic sludge blanket, A-TRAP = A-stage of the AB-process, SBR = sequencing batch reactor, TF = trickling filter)*

Black water The excess sludge from the UASB reactor operated on black water can be used as **organic fertilizer** after appropriate post-treatment to further disinfect the sludge. In this way the carbon cycle can be closed between households and agriculture. Calcium phosphate granules produced in the sludge bed of the UASB reactor represent an alternative approach to **phosphate** recovery. Further research is needed to render all of the phosphate in black water as pure calcium phosphate granules. The effluent from the UASB reactor can be discharged after appropriate post-treatment to remove the nitrogen and remaining organic matter. **Energy** can be recovered as methane from the UASB reactor, and addition of kitchen refuse can be used to increase the methane production, provided that the level of heavy metals in the sludge is not increased. Further research should also investigate the influence of kitchen refuse addition on the calcium phosphate granulation process in the sludge bed. By decreasing the flushing volume for the vacuum collection of black water from 6 to 1.5 L/cap/d, the energy consumption of black water treatment can be decreased by 35–55%, as presented in Chapter 2. Furthermore, lower flushing volume decreases the heavy metal loading from tap water, in particular As, Cu and Pb (Chapter 3), and the 75% reduction in the flushing volume decreases the heavy metal loading of black water by 39% for As, 23% for Cu and 21% for Pb. Also, the lower the dilution, the more phosphate is retained in the sludge bed of the UASB reactor [43], possibly improving the calcium phosphate granulation process. Further investigations should be done on the practical implications of lower flushing volume of black water vacuum collection.

Grey water Two different treatment options are recommended for grey water. To utilize the **energy** potential, grey water can be bioflocculated in an A-trap, and the produced sludge can be digested for methane production. To ensure the purity of black water sludge, grey water sludge should be treated in a separate anaerobic digestion unit. However, this increases the complexity of the treatment concept, and further investigations should be done on the practical and economical feasibility of two separate digestion units. Alternatively, grey water can be treated in a SBR producing stabilized sludge at lower quantities. This system, however, requires higher aeration energy without the recovery of methane compared to the combination of A-trap and anaerobic digestion. In terms of **water** recovery, the use of a SBR is more beneficial as the effluent can be reused according to the standards for unrestricted urban reuse after post-treatment with a trickling filter (TF) (Chapter 2).

Summary

Source-separation and on-site treatment of domestic wastewater presents a great potential for resource recovery. In this thesis the integration of treatment systems for black and grey water was investigated to improve resource recovery within source-separated sanitation concepts, with a special focus on phosphate and organic fertilizer recovery from vacuum collected black water. **Chapter 1** describes the current state of agricultural soil in terms of declining soil fertility, and the problems related to the use of artificial phosphate fertilizers. The organic matter content of soil is a key factor determining its nutrient-holding capacity, and the supply of both organic and mineral nutrients is therefore essential to sustain food production. A major portion of the nutrients leaving agriculture is in the food products, and human excreta contribute a significant fraction of the nitrogen and phosphorus flows in society. By source-separation of black water, external input of heavy metals, such as industrial effluents, surface run-offs and grey water can be eliminated, and organic carbon and nutrients returned back to the soil/food cycle, reducing the need for artificial fertilizers. Phosphate fertilizers are predominantly sourced from phosphate rock – a resource that is declining in both quantity and quality. Phosphate recovery locally from waste streams is therefore a prerequisite for global food security and sustainability. Black water/feces and urine with lower levels of heavy metals and micropollutants compared to sewage and manure are ideal sources for phosphate recovery.

Optimal resource recovery is dependent on the selection of existing treatment processes and the local environmental conditions. The integration of different treatment processes at different levels of source-separation for energy, water and nutrient recov-

ery was investigated in **Chapter 2** using a mass and energy balance-based model. The model was constructed from literature data to compare energy and water balance, nutrient recovery, chemical use, effluent quality and land area requirement in 4 different sanitation concepts: (1) centralized; (2) centralized with source-separation of urine; (3) source-separation of black water, kitchen refuse and grey water; and (4) source-separation of urine, feces, kitchen refuse and grey water. The highest primary energy consumption of 914 MJ/cap/year was attained within the centralized sanitation concept, and the lowest primary energy consumption of 437 MJ/cap/year was attained within source-separation of urine, feces, kitchen refuse and grey water. Grey water bioflocculation and subsequent grey water sludge co-digestion decreased the primary energy consumption, but was not energetically favorable to couple with grey water effluent reuse. Source-separation of urine improved the energy balance, nutrient recovery and effluent quality, but required larger land area and higher chemical use in the centralized concept. By lowering the flushing volume from 6 to 1.5 L/cap/d for vacuum collection of black water, the energy consumption of black water treatment can be decreased by 35–55%, resulting in the lowest primary energy consumption of 156 MJ/cap/year from all the sanitation concepts.

Heavy metal content of sewage sludge is currently the most significant factor limiting its reuse in agriculture within the European Union. In the Netherlands most of the produced sewage sludge is incinerated, mineralizing the organic carbon into the atmosphere rather than returning it back to the soil. Source-separation and vacuum collection of black water excludes external heavy metal inputs, such as industrial effluents, surface run-offs, grey water and tap water, and is therefore a more favorable source for resource recovery. However, the soil application of black water sludge is prohibited in the Netherlands due to elevated Cu and Zn concentrations. In **Chapter 3** a full heavy metal mass balance of black water was drawn based on literature data with additional control samples analyzed within this study. The contribution of different black water components to the total heavy metal loading of black water was determined, and the primary origin of heavy metals in the black water components was investigated. The results from this study showed that feces is the main contributor to the heavy metal loading in vacuum collected black water (52%–84%), while in sewage the contribution of feces is less than 10%. The use of the same guidelines for sewage and black water can be therefore argued, since black water is mainly characterized by human originated content (feces and urine) while sewage is a mixture of industrial and domestic wastewater, and rainwater. To distinguish these two streams in the sludge reuse regulation, a control parameter should be implemented, such as the Hg and Pb content that is significantly higher in sewage sludge compared to black water sludge (from 50– to 200–fold). The heavy metals in feces and urine are primarily from dietary sources, and promotion of the soil application of black water sludge over livestock manure and artificial fertilizers could further reduce the heavy metal content in the soil/food cycle.

The concentrated streams of black water and kitchen refuse are considered the main sources for energy recovery within source-separated sanitation concepts. To further improve energy recovery, the organic matter present in grey water can be concentrated in a bioflocculation unit and subsequently digested with black water. In **Chapter 4** the potential of combining anaerobic grey water sludge treatment with black water in an upflow anaerobic sludge blanket (UASB) reactor was investigated. Black water and the mixture of black water and grey water sludge were compared in terms of biochemical methane potential (BMP), UASB reactor performance, COD mass balance and methanization. Grey water sludge treatment with black water increased the energy recovery by 23% in the UASB reactor compared to black water treatment, not only due to additional organic fraction, but also by increasing the methanization. The increase in the energy recovery can cover the increased heat demand of the UASB reactor and the electricity demand of the grey water bioflocculation system with a surplus of 0.7 kWh/cap/y electricity and 14 MJ/cap/y heat. However, grey water sludge introduced more heavy metals in the excess sludge of the UASB reactor, and might therefore hinder its soil application.

Recovery of phosphate from wastewater in the form of struvite is widely applied, mostly due to its spontaneous precipitation process and the co-precipitation of ammonium, resulting in a product that can be reused as a slow release fertilizer in agriculture. However, recovered phosphate product needs to be certified in order to be used as a fertilizer, and the reuse of struvite in agriculture is therefore limited. Recovery of calcium phosphate is more beneficial as it has the effective composition of phosphate rock, and can be therefore used as raw material in fertilizer industry. In **Chapter 5** a novel approach was introduced to recover phosphate by precipitation of calcium phosphate granules in anaerobic treatment of black water. In this approach, phosphate and energy can be simultaneously recovered in one anaerobic reactor. The granules formed in the upflow anaerobic sludge blanket (UASB) reactor at lab– and demonstration–scale were analyzed for chemical composition and mineralogy by Inductively Coupled Plasma–Atomic Emission Spectroscopy (ICP–AES) and direct spectral analysis. The granules had a diameter of 1–2 mm, organic content of 33 wt%, and phosphorus content of 11–13 wt%. Three calcium phosphate phases were identified in the granules: hydroxyapatite, calcium phosphate hydrate and carbonated hydroxyapatite. Without any addition of chemicals, 7 gP/person/year can be recovered with the calcium phosphate granules, representing 2% of the incoming phosphorus in the UASB reactor. As the heavy metal content was lower compared to other phosphorus recovery products, phosphate rock and phosphorus fertilizer, the calcium phosphate granules could be considered as a new phosphorus product.

In **Chapter 6** phosphate and organic fertilizer recovery from black water is further discussed, and a sanitation concept is proposed based on the knowledge gathered in this thesis. In order to close the soil/food heavy metal cycle, external heavy metal sources, such as manure and artificial fertilizers should be included in the heavy metal regulation. Furthermore, to retain the purity of black water sludge, emphasis should be laid on the excess sludge quality when co-digesting black water with kitchen refuse or grey water sludge. A pretreatment step to disinfect black water sludge prior to soil application is recommended, and different options, such as co-composting with garden waste and hyper-thermophilic treatment are proposed. Cooperation between the different stakeholder groups of building companies, agricultural sector, water boards, and the government is needed for successful introduction of new fertilizer products in agriculture. The goal of optimal phosphate recovery is production of high purity product in an energy efficient reactor configuration with a minimum addition of chemicals. The occurrence of high purity calcium phosphate granules in this study can be attributed to the simultaneous influence of chemical, physical and biological factors in the sludge bed of the UASB reactor operated on black water. To optimize the calcium phosphate granulation, a small amount of additional calcium together with seed material can be dosed using a fluidized bed reactor configuration. In addition, selective use of micro-organisms can increase the calcium phosphate granulation by enhanced metabolic activity. Finally, a sanitation concept is proposed in which energy, phosphate in the form of calcium phosphate granules, and organic fertilizer is recovered from vacuum collected black water using a lower flushing volume. Grey water is treated separately to recover water, and alternatively to recover energy in another anaerobic treatment unit.

Samenvatting

Nieuwe sanitatie, ook wel brongescheiden sanitatie genoemd, heeft een groot potentieel om belangrijke grondstoffen uit huishoudelijk afvalwater terug te winnen. In dit proefschrift is een gentegreerd behandelingsconcept voor zwart en grijs water onderzocht, met als doel maximale terugwinning van grondstoffen. De focus ligt hierbij vooral op de terugwinning van fosfaat en organische meststoffen uit met vacumtoiletten ingezameld zwart water. **Hoofdstuk 1** beschrijft de huidige situatie rond de afnemende bodemvruchtbaarheid van landbouwgrond en de problemen die verbonden zijn aan het gebruik van kunstmatige fosfaatmeststoffen. Het organische stof gehalte van de bodem is bepalend voor de capaciteit van de bodem om voedingsstoffen vast te houden. Om de voedselproductie in stand te houden, is toevoeging van zowel organische stof als minerale nutriënten essentieel. Een groot deel van de nutriënten in de grond komt terecht in het voedsel en wordt door de mens, via de urine en fecaliën, weer in het milieu teruggebracht. Door gescheiden inzameling van zwart water, worden extra emissies van zware metalen afkomstig van industrieel afvalwater, vervuild regenwater en grijs water verminderd of voorkomen. Hierdoor kan het slib, afkomstig uit gescheiden verwerking, als meststof gebruikt worden waardoor de organische koolstof en nutriënten weer op het land en in de voedselcyclus gebracht worden en het gebruik van kunstmatige meststoffen overbodig wordt. Fosfaatmeststoffen worden voornamelijk gewonnen uit fosfaaterts, een bron die steeds schaarser wordt en afneemt in kwaliteit Om de wereldvoedselproductie op peil te houden wordt terugwinning van fosfaat uit reststromen noodzakelijk. Zwart water/fecaliën en urine bevatten minder zware metalen en microverontreinigingen vergeleken met rioolwater en mest en zijn daarom ideale bronnen om fosfaat uit terug te winnen.

Afhankelijk van het juiste behandelingsproces en de lokale milieuomstandigheden, vindt een optimale terugwinning van grondstoffen plaats. In **Hoofdstuk 2** is op een modelmatige wijze de integratie van verschillende behandelingsprocessen gericht op terugwinning van energie, water en nutriënten onderzocht. Het model is gebaseerd op literatuurdata en vergelijkt de water– en energiebalans, de nutriëntenterugwinning, het chemicaliëngebruik, de effluentkwaliteit en het benodigde grondoppervlak (footprint) voor 4 verschillende sanitatieconcepten: (1) gecentraliseerd, (2) gecentraliseerd met separate inzameling van urine, (3) brongescheiden inzameling van zwart water, groente– en fruitafval en grijs water, en (4) brongescheiden inzameling van urine, fecaliën, groente– en fruitafval en grijs water. Het centrale sanitatieconcept (1) heeft het hoogste primaire energieverbruik (914 MJ/persoon/jaar). Het sanitatieconcept bestaande uit brongescheiden inzameling van urine, fecaliën, groente– en fruitafval en grijs water (4) heeft het laagste primaire energieverbruik (437 MJ/persoon/jaar). Het primaire energieverbruik neemt af bij grijs water bioflocculatie in combinatie met co–vergisting van grijs water slib, maar de koppeling met hergebruik van grijs water effluent werkt negatief uit op het energieverbruik. De energiebalans, het terugwinnen van nutriënten en de kwaliteit van het effluent verbeteren bij brongescheiden inzameling van urine. Wel vereist dit concept (2) meer grondoppervlak en meer chemicaliën dan het centrale concept (1). Verlaging van het spoelvolume van vacuumtoiletten van 6 naar 1,5 L/persoon/dag, leidt tot een verlaging in energieverbruik van de zwart waterbehandeling van 35–55%, resulterend in het laagste primaire energieverbruik van 156 MJ/persoon/jaar.

Het gehalte aan zware metalen in communaal zuiveringsslib verhindert op dit moment binnen de Europese Unie de toepassing van dit slib als meststof in de landbouw. In Nederland wordt zuiveringsslib met name verbrand, waarbij de organische koolstof gemineraliseerd wordt en in de atmosfeer terecht komt in plaats van in de bodem. Het slib dat als restproduct vrijkomt bij gescheiden inzameling van zwart water met vacuumtoiletten, bevat aanzienlijk minder zware metalen dan communaal zuiveringsslib. De te hoge concentraties aan koper en zink staan evenwel toepassing als meststof in de landbouw in Nederland nog in de weg. In **Hoofdstuk 3** wordt een volledige massabalans voor zware metalen in zwart water gepresenteerd op basis van literatuurgegevens, aangevuld met controlemonsters die in het kader van dit onderzoek zijn geanalyseerd. Hieruit blijkt dat fecaliën de grootste bijdrage leveren aan het gehalte zware metalen in met vacumtoiletten ingezameld zwart water (52%–84%), in rioolwater bedraagt dit aandeel vanuit fecaliën minder dan 10%. Het gebruik van dezelfde richtlijnen voor toepassing van zuiveringsslib en slib afkomstig van zwart waterbehandeling in de landbouw is daarom betwistbaar, omdat zwart water voornamelijk producten van menselijke oorsprong (fecaliën en urine) bevat, terwijl rioolwater een mengsel is van industrieel en huishoudelijk afvalwater en regenwater. In de regelgeving voor hergebruik zou een onderscheid tussen deze twee stromen terecht zijn door het invoeren van een controleparameter, zoals het kwik– en loodgehalte. Dit is in zuiveringsslib aanzienlijk hoger dan in zwart water slib (factor 50 tot 200 hoger). De zware metalen in fecaliën en urine zijn voornamelijk afkomstig uit voedsel en toepassing van zwart water slib als meststof in plaats van dierlijke mest en kunstmest leidt tot een verlaging van het zware metalen gehalte in de bodem en voedselketen.

De geconcentreerde stromen zwart water en groente– en fruitafval zijn de belangrijkste bronnen voor energie terugwinning binnen brongescheiden sanitatie. De energieopbrengst kan verhoogd worden door de organische stof aanwezig in grijs water te concentreren in een bioflocculatie unit en daarna te vergisten samen met zwart water. De potentie van dit concept is in **Hoofdstuk 4** onderzocht. Zwart water en een mengsel van zwart water en grijs water slib is met elkaar vergeleken op de volgende parameters: de potentiële methaanproductie (BMP), werking UASB (upflow anaerobic sludge blanket) reactor, CZV–massabalans en daadwerkelijke methaanproductie. Door grijs water slib samen met zwart water te behandelen in een UASB reactor, neemt de energieproductie toe met 23%, vergeleken met alleen zwart water behandeling. Dit komt niet alleen door de extra vracht aan organische stof maar ook door de toegenomen methaanproductie. De toename in energieopbrengst dekt de extra benodigde hoeveelheid warmte voor de UASB reactor en het elektriciteitsverbruik van de grijs waterbehandeling (bioflocculatie) waarbij een surplus resteert van 0,7 kWh/persoon/jaar aan elektriciteit en 14 MJ/persoon/jaar aan warmte. Toevoeging van grijs water slib resulteert wel in een hoger gehalte aan zware metalen in het UASB–slib en kan daardoor toepassing als meststof belemmeren.

Terugwinning van fosfaat uit afvalwater in de vorm van struviet wordt al breed toegepast, vooral vanwege het spontane precipitatieproces en de coprecipitatie van ammonium. Doordat de voedingsstoffen langzaam vrijkomen resulteert dit in een langwerkende meststof. Om het product als meststof toe te passen dient het gecertificeerd te zijn en om die reden is toepassing van struviet nog gelimiteerd. Terugwinning van calciumfosfaat verdient de voorkeur omdat de samenstelling hiervan vergelijkbaar is met die van fosfaaterts. Calciumfosfaat kan als grondstof gebruikt wordt in de kunstmestindustrie. In **Hoofdstuk 5** wordt een nieuwe benadering gentroduceerd om tijdens de anaerobe behandeling van zwart water, fosfaat terug te winnen in de vorm van calciumfosfaat korrels. Op deze manier kan in een anaerobe reactor zowel fosfaat als energie teruggewonnen worden. De chemische en mineralogische samenstelling van de op lab– en demonstratieschaal geproduceerde korrels is bepaald door middel van Inductively Coupled PlasmaAtomic Emission Spectroscopy (ICP-AES) en directe spectrale analyse. De korrels hebben een diameter van 1–2 mm, een organisch stofgehalte van 33 gewicht% en een fosfaatgehalte van 11–13 gewicht%. Drie vormen van calciumfosfaat zijn gedentificeerd in de korrels: hydroxyapatite, calcium fosfaat hydraat en carbonated hydroxyapatite. Zonder enige toevoeging van chemicaliën kan 7 gP/persoon/jaar worden teruggewonnen in calciumfosfaat korrels, wat overeenkomt met 2% van de fosfaatvracht in de UASB reactor. Omdat het gehalte aan zware metalen lager is in vergelijking met andere fosfaatproducten, zoals fosfaaterts en fosfaatkunstmest, kunnen de calciumfosfaat korrels gezien worden als een nieuw fosfor product.

In **Hoofdstuk 6** vindt een verdere discussie plaats over de terugwinning van fosfaat en organische meststoffen uit zwart water en wordt een sanitatieconcept voorgesteld gebaseerd op de kennis vergaard dit proefschrift. Om de zware metalen kringloop in bodem/voedsel te sluiten moeten externe bronnen van zware metalen, zoals dierlijke mest en kunstmest, meegenomen worden in de regelgeving. Om zo zuiver mogelijk slib te produceren, dient de kwaliteit van extra toe te voegen slib goed in de gaten gehouden worden, wanneer zwart water in combinatie met groente– en fruitafval of grijs water slib wordt vergist. Een voorbehandelingstap voor zwart water slib bestaande uit desinfectie, wordt aanbevolen voorafgaand aan de toepassing als meststof en verschillende opties zoals co–compostering met tuinafval en hyper thermofiele behandeling worden voorgesteld. Een goede samenwerking tussen de verschillende stakeholders zoals de bouwsector, de agrarische sector, sanitairleveranciers, waterschappen en overheden is noodzakelijk voor een succesvolle introductie van nieuwe meststoffen in de landbouw. Het doel van optimale fosfaatterugwinning is de productie van een zuiver product in een energie–efficiënte reactorconfiguratie met een minimale toevoeging van chemicaliën. Het ontstaan van zuivere calciumfosfaat korrels in deze studie wordt toegeschreven aan de gelijktijdige invloed van chemische, fysische en biologische factoren in het slib bed van de UASB reactor. De vorming van calciumfosfaat korrels kan geoptimaliseerd worden door een kleine hoeveelheid calcium in combinatie met entmateriaal toe te voegen in een fluidized bed reactor. Daarnaast kan bij een selectief gebruik van micro–organismen met een verhoogde metabolische activiteit de korrelproductie verhoogd worden. Het proefschrift sluit af met een sanitatieconcept waarbij energie, fosfaat in de vorm van calciumfosfaat korrels en organische meststof wordt teruggewonnen uit vacum ingezameld zwart water. Grijs water wordt apart behandeld teneinde het water her te gebruiken of energie terug te winnen in een aparte anaerobe behandelingsstap.

Bibliography

[1] L. Abu-Ghunmi, G. Zeeman, M. Fayyad, and J. B. van Lier. Grey water treatment systems: a review. *Critical reviews in environmental science and technology*, 41(7):657–698

[2] M. Adamsson. Potential use of human urine by greenhouse culturing of microalgae (scenedesmus acuminatus), zooplankton (daphnia magna) and tomatoes (lycopersicon). *Ecological Engineering*, 16:243–254, 2000.

[3] O. M. Ağdağ and D. T. Sponza. Co-digestion of industrial sludge with municipal solid wastes in anaerobic simulated landfilling reactors. *Process Biochemistry*, 40(5):1871–1879, 2005.

[4] R. Alvarez, L. A Evans, P. J Milham, and M. A Wilson. Effects of humic material on the precipitation of calcium phosphate. *Geoderma*, 118(3):245–260, 2004.

[5] I. Angelidaki and W. Sanders. Assessment of the anaerobic biodegradability of macropollutants. *Reviews in Environmental Science and Biotechnology*, 3(2): 117–129, 2004.

[6] APHA. *Standard Methods for the Examination of Waste and Wastewater.* American Public Health Association, New York, 1998.

[7] P. Apostoli. Elements in environmental and occupational medicine. *Journal of Chromatography B*, 778(1):63–97, 2002.

[8] K. Ashley, D Cordell, and D Mavinic. A brief history of phosphorus: From the philosopher's stone to nutrient recovery and reuse. *Chemosphere*, 84(6), 2011.

[9] L. M. Avery, R. A. D. Frazer-Williams, G. Winward, C. Shirley-Smith, S. Liu, F. A. Memon, and B. Jefferson. Constructed wetlands for grey water treatment. *Ecohydrology & Hydrobiology*, 7(3-4):191–200, 2007.

[10] W. Becker, L. Jorhem, B. Sundström, and K. Petersson Grawé. Contents of mineral elements in swedish market basket diets. *Journal of Food Composition and Analysis*, 24(2):279–287, 2011.

[11] E. Benetto, D. Nguyen, T. Lohmann, B. Schmitt, and P. Schosseler. Life cycle assessment of ecological sanitation system for small-scale wastewater treatment. *Science of The Total Environment*, 407(5):1506–1516, 2009.

[12] U. Berg. P-roc–ein verfahren zur p–rückgewinnung aus abwässern durch abscheidung von calciumphosphat–mineralphasen an tobermorit. In *75. Darmstädter Seminar–Rückgewinnung von Phosphor aus Abwasser und Klärschlamm, Schriftenreihe WAR, Nr. 167*, volume 1679, Darmstadt, Germany, 2005.

[13] G. H. Biego, M. Joyeux, P. Hartemann, and G. Debry. Daily intake of essential minerals and metallic micropollutants from foods in france. *Science of The Total Environment*, 217(1-2):27–36, 1998.

[14] E. Bligh and W. Dyer. A rapid method of total lipid extraction and purification. *Canadian journal of biochemistry and physiology*, 37(8):911–917 0576–5544, 1959.

[15] A. Bocio, M. Nadal, and J. L. Domingo. Human exposure to metals through the diet in tarragona, spain. *Biological trace element research*, 104(3):193–201, 2005.

[16] B. Böhnke. Energieminimierung durch das adsorptions-belebungsverfahren. *Gewässerschutz Wasser Abwasser*, 49, 1981.

[17] BOOM. Manual of environmental policy (in dutch), 2008. Accessed on: 15 October 2013. URL `http://www.eu-milieubeleid.nl/ch05s10.html`.

[18] M. G. Bos, W. H. Diemont, and A. Verhagen. Sustainable peat supply chain: Report of the ad hoc working group enhancing the sustainability of the peat supply chain for the dutch horticulture. Alterra, Wageningen University, 2011.

[19] A. L Boskey and A. S Posner. Conversion of amorphous calcium phosphate to microcrystalline hydroxyapatite. a pH-dependent, solution-mediated, solid-solid conversion. *The Journal of Physical Chemistry*, 77(19):2313–2317, 1973.

[20] Z. Bradford-Hartke, P. Lant, and G. Leslie. Phosphorus recovery from centralised municipal water recycling plants. *Chemical Engineering Research and Design*, 90(1), 2012.

[21] M. Braungart, W. McDonough, and A. Bollinger. Cradle-to-cradle design: creating healthy emissions – a strategy for eco-effective product and system design. *Journal of Cleaner Production*, 15(13-14):1337–1348, 2007.

[22] A. Britton, R. Prasad, B. Balzer, and L. Cubbage. *Pilot testing and economic evaluation of struvite recovery from dewatering centrate at HRSD's Nansemond WWTP.* 2009.

[23] H. Brix and C. A. Arias. The use of vertical flow constructed wetlands for on-site treatment of domestic wastewater: New danish guidelines. *Ecological Engineering*, 25:491–500, 2005.

[24] J. P. Buchet, R. Lauwerys, A. Vandevoorde, and J. M. Pycke. Oral daily intake of cadmium, lead, manganese, copper, chromium, mercury, calcium, zinc and arsenic in belgium: a duplicate meal study. *Food and chemical toxicology*, 21 (1):19–24, 1983.

[25] V. W. Bunker, M. S. Lawson, H. T. Delves, and B. E. Clayton. The intake and excretion of lead and cadmium by the elderly. *The American Journal of Clinical Nutrition*, 39(5):803–808, 1984.

[26] X. Cao and W. Harris. Carbonate and magnesium interactive effect on calcium phosphate precipitation. *Environmental Science & Technology*, 42(2):436–442, 2008.

[27] CBS. Urban wastewater treatment per province and river basin district, 2011. Accessed on: 15 January 2014. URL `www.cbs.nl`.

[28] IFDC-International Fertilizer Development Center, 2014. Accessed on: 13 May 2014. URL `http://www.ifdc.org`.

[29] F. Claeys-Thoreau, L. Thiessen, P. Bruaux, G. Ducoffre, and G. Verduyn. Assessment and comparison of human exposure to lead between belgium, malta, mexico and sweden. *International archives of occupational and environmental health*, 59(1):31–41, 1987.

[30] A. J. Cohen and F. J. C. Roe. Review of lead toxicology relevant to the safety assessment of lead acetate as a hair colouring. *Food and chemical toxicology*, 29 (7):485–507, 1991.

[31] S. D. W. Comber and A. M. Gunn. Heavy metals entering sewage-treatment works from domestic sources. *Water and Environment Journal*, 10(2):137–142, 1996.

[32] C. Combes and C. Rey. Amorphous calcium phosphates: Synthesis, properties and uses in biomaterials. *Acta Biomaterialia*, 6(9):3362–3378, 2010.

[33] E. Comino, V. A. Riggio, and M. Rosso. Biogas production by anaerobic co-digestion of cattle slurry and cheese whey. *Bioresource Technology*, 114:46–53 0960–8524, 2012.

[34] Commission of the European Communities. The white paper of RES. Brussels, 1997.

[35] D. Cordell, J. O Drangert, and S. White. The story of phosphorus: Global food security and food for thought. *Global Environmental Change*, 19(2):292–305, 2009.

[36] P. Cornel and C. Schaum. Phosphorus recovery from wastewater: needs, technologies and costs. *Water Science & Technology*, 59(6):1069, 2009.

[37] E. A. Davidson and I. A. Janssens. Temperature sensitivity of soil carbon decomposition and feedbacks to climate change. *Nature*, 440(7081):165–173, 2006.

[38] Å. Davidsson, C. Lövstedt, J. la Cour Jansen, C. Gruvberger, and H. Aspegren. Co-digestion of grease trap sludge and sewage sludge. *Waste Management*, 28 (6):986–992 0956–053X, 2008.

[39] J.C. Dawson and P. Smith. Carbon losses from soil and its consequences for land-use management. *Science of The Total Environment*, 382(2-3):165–190, 2007.

[40] A.J. de Buck, W. van Dijk, J.C. van Middelkoop, A.L. Smit, H. van Reuler, and A. Evers. *Agricultural scenarios to reduce the national phosphorus surplus in the Netherlands.* Applied Plant Research (Praktijkonderzoek Plant & Omgeving), part of Wageningen UR Business Unit AGV, 2012.

[41] M. S. de Graaff. *Resource recovery from black water.* PhD thesis, Wageningen University, 2010.

[42] M. S. de Graaff, H. Temmink, G. Zeeman, and C. J. N. Buisman. Anaerobic treatment of concentrated black water in a UASB reactor at a short HRT. *Water*, 2(1):101–119, 2010.

[43] M. S. de Graaff, H. Temmink, G. Zeeman, and C. J. N. Buisman. Energy and phosphorus recovery from black water. *Water Science & Technology*, 63(11): 2759, 2011.

[44] M. S. de Graaff, N. M. Vieno, K. Kujawa-Roeleveld, G. Zeeman, H. Temmink, and C. J. N. Buisman. Fate of hormones and pharmaceuticals during combined anaerobic treatment and nitrogen removal by partial nitritation-anammox in vacuum collected black water. *Water research*, 45(1):375–383, 2011.

[45] DeSaH B.V. Reactor performance of the AB-process and the UASB reactor at the DeSaH demonstration site, 2010.

[46] B. Dittrich and R. Klose. Schwermetalle in düngemitteln: Schriftenreihe der sächsischen landesanstalt für landwirtschaft, 2008.

[47] W. Van Dokkum, R. H. De Vos, T. H. Muys, and J. A. Wesstra. Minerals and trace elements in total diets in the netherlands. *British Journal of Nutrition*, 61(01):7–15, 1989.

[48] S. V. Dorozhkin and M. Epple. Biological and medical significance of calcium phosphates. *Angewandte Chemie International Edition*, 41(17):3130–3146, 2002.

[49] H. L. Dorussen. Ontwerp awzi dokhaven, 1980.

[50] J. D. Doyle and S. A. Parsons. Struvite formation, control and recovery. *Water Research*, 36(16):3925–3940, 2002.

[51] J. Driver, D. Lijmbach, and I. Steen. Why recover phosphorus for recycling, and how? *Environmental technology*, 20(7):651–662, 1999.

[52] T. A. Elmitwalli, J. Soellner, A. de Keizer, H. Bruning, G. Zeeman, and G. Lettinga. Biodegradability and change of physical characteristics of particles during anaerobic digestion of domestic sewage. *Water Research*, 35(5):1311–1317, 2001.

[53] European Commission. Council Directive 86/278/EEC on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture, 1986. Accessed on: 10 October 2013. URL `http://ec.europa.eu/environment/waste/sludge`.

[54] European Commission. Directive 2006/12/EC of european parliament and of the council of 5 april 2006 on waste. Brussels, 2006.

[55] European Commission. Sewage sludge, 2012. Accessed on: 18 October 2013. URL `http://ec.europa.eu/environment/waste/sludge`.

[56] European Commission. EU Water Framework Directive 91/271/EEC, 2013. Accessed on: 3 February 2013. URL `http://ec.europa.eu/environment/water/water-framework`.

[57] M. Franz. Phosphate fertilizer from sewage sludge ash (SSA). *Waste Management*, 28(10):1809–1818, 2008.

[58] J. Frijns, M. Mulder, and J. Roorda. Op weg naar een klimaatneutrale waterketen (in dutch). Utrecht, the Netherlands, 2008.

[59] K. Fujie, H. E. Bravo, and H. Kubota. Operational design and power economy of a rotating biological contactor. *Water Research*, 17(9):1153–1162, 1983.

[60] J. L. García-Morales, E. Nebot, L. I. Romero, and D. Sales. Comparison between acidogenic and methanogenic inhibition caused by linear alkylbenzenesulfonate (LAS). *Chemical and biochemical engineering quarterly*, 15(1):13–20, 2001.

[61] M. R. Gaterell, R. Gay, R. Wilson, R. J. Gochin, and J. N. Lester. An economic and environmental evaluation of the opportunities for substituting phosphorus recovered from wastewater treatment works in existing UK fertiliser markets. *Environmental Technology*, 21(9):1067–1084, 2000.

[62] R. George. *The Big Necessity: The unmentionable world of human waste and why it matters.* Macmillan, 2008.

[63] A. Giesen, R. de Boer, and A. Gaillard. *Practical Experience with Recovery of Phosphates from Wastewater using Crystallization Technology. 75*, volume 167. 2005.

[64] GMB. SaNiPhos. Obheusden, Netherlands, 2010. Accessed on: 13 June 2014. URL `http://www.saniphos.eu/`.

[65] A. Gobin, P. Campling, L. Janssen, N. Desmet, H. van Delden, J. Hurkens, P. Lavelle, and S. Berman. Soil organic matter management across the EU—best practices, constraints and tradeoffs. *Final report for the European Commission's DG Environment*, 2011.

[66] D. Gompertz, C. Courage, P. Illing, and M. Taylor. IEH report on factors affecting the absorption of toxic metals from the diet. Leicester, England, 1998.

[67] P. Grandjean, O. Andersen, and G. D. Nielsen. *Biological indicators for the assessment of human exposure to industrial chemicals.* Commission of the European Communities, 1988.

[68] J. S. Guest, S. J. Skerlos, J. L. Barnard, M. B. Beck, G. T. Daigger, H. Hilger, S. J. Jackson, K. Karvazy, L. Kelly, L. Macpherson, J. R. Mihelcic, A. Pramanik, L. Raskin, M. C. M. Van Loosdrecht, D. Yeh, and N. G. Love. A new planning and design paradigm to achieve sustainable resource recovery from wastewater. *Environmental Science & Technology*, 43(16):6126–6130, 2009.

[69] J. F. Hair, R. E. Anderson, R. L. Tatham, and W. C. Black. Multivariate analysis. *Englewood: Prentice Hall International*, 1998.

[70] W. G. Harris, A. C. Wilkie, X. Cao, and R. Sirengo. Bench-scale recovery of phosphorus from flushed dairy manure wastewater. *Bioresource Technology*, 99 (8):3036–3043, 2008.

[71] D. Hellström. Exergy analysis: A comparison of source separation systems and conventional treatment systems. *Water environment research*, 71(7):1354–1363, 1999.

[72] J. G. Hering, T. D. Waite, R. G. Luthy, J. E. Drewes, and D. L. Sedlak. A changing framework for urban water systems. *Environmental Science & Technology*, 47(19):10721–10726, 2013.

[73] L. Hernández Leal. *Removal of micropollutants from grey water: combining biological and physical/chemical processes*. PhD thesis, Wageningen University, 2010.

[74] L. Hernández Leal, H. Temmink, G. Zeeman, and C. J. N. Buisman. Bioflocculation of grey water for improved energy recovery within decentralized sanitation concepts. *Bioresource technology*, 2010.

[75] L. Hernández Leal, H. Temmink, G. Zeeman, and C. J.N Buisman. Comparison of three systems for biological greywater treatment. *Water*, 2(2):155–169, 2010.

[76] L. Hernández Leal, H. Temmink, G. Zeeman, and C.J.N. Buisman. Characterization and anaerobic biodegradability of grey water. *Desalination*, 270(1-3): 111–115, 2011.

[77] J. B. Holm-Nielsen, T. Al Seadi, and P. Oleskowicz-Popiel. The future of anaerobic digestion and biogas utilization. *Bioresource Technology*, 100(22):5478–5484 0960–8524, 2009.

[78] G. Houillon and O. Jolliet. Life cycle assessment of processes for the treatment of wastewater urban sludge: energy and global warming analysis. *Journal of cleaner production*, 13(3):287–299, 2005.

[79] L. W. Hulshoff Pol, S. I. de Castro Lopes, G. Lettinga, and P. N. L. Lens. Anaerobic sludge granulation. *Water Research*, 38(6):1376–1389, 2004.

[80] IFPRI. Green revolution: Curse or blessing? *International Food Policy Research Institute (IFPRI)*, 2002.

[81] E. Jarosewich. Smithsonian microbeam standards. *Journal of Research of the National Institute of Standards and Technology*, 107(6):681–685, 2002.

[82] J. C. Jenkins. *The humanure handbook: A guide to composting human manure.* Joseph Jenkins, Inc., Grove City, PA 16127 USA, 3rd edition, 2005.

[83] Z. Ji, G. Chen, and Y. Chen. Effects of waste activated sludge and surfactant addition on primary sludge hydrolysis and short-chain fatty acids accumulation. *Bioresource technology*, 101(10):3457–3462, 2010.

[84] H. Jönsson and B Vinnerås. Closing the loop: Recycling nutrients to agriculture. In T. A. Larsen, K. M. Udert, and J. Lienert, editors, *Source Separation and Decentralization for wastewater Management*, pages 163–176. IWA Publishing, London, UK, 2013.

[85] Chris Kaper. Duurzame fosfaatverwijdering op rwzi geestmerambacht. Technical report, Hogeschool Utrecht, Milieukunde duaal, 2009.

[86] I. J. Karassik, J. P. Messina, P. Cooper, and C. C. Heald, editors. *Pump Handbook.* McGraw-Hill, 3rd edition, 2001.

[87] A. Kelessidis and A. S. Stasinakis. Comparative study of the methods used for treatment and final disposal of sewage sludge in european countries. *Waste management*, 32(6), 2012.

[88] H. Kirchmann and S. Pettersson. Human urine-chemical composition and fertilizer use efficiency. *Fertilizer research*, 40(2):149–154, 1994.

[89] KNMI. Annual report, 2010. Accessed on: 3 June 2013. URL `http://www.knmi.nl/`.

[90] M. Koch and W. Rotard. On the contribution of background sources to the heavy metal content of municipal sewage sludge. *Water science and technology*, 43(2):67–74, 2001.

[91] K. Kujawa. *Anaerobic treatment of concentrated wastewater in DESAR concepts.* STOWA, 2005.

[92] K. Kujawa-Roeleveld and G. Zeeman. Anaerobic treatment in decentralised and source-separation-based sanitation concepts. *Reviews in Environmental Science and Bio/Technology*, 5(1):115–139, 2006.

[93] K. Kujawa-Roeleveld, T. Elmitwalli, A. Gaillard, M. van Leeuwen, and G. Zeeman. Co-digestion of concentrated black water and kitchen refuse in an accumulation system within the desar (decentralized sanitation and reuse) concept. *Water Science and Technology*, 48(4):121–128, 2003.

[94] K. Kujawa-Roeleveld, J. Weijma, and T. Nanninga. Nieuwe sanitatie op wijkniveau: Resultaten en ervaringen demo-site sneek, 2012.

[95] P. Kuntke. *Nutrient and energy recovery from urine*. PhD thesis, Wageningen University, 2013.

[96] P. Kuntke, K.M. Śmiech, H. Bruning, G. Zeeman, M. Saakes, T.H.J.A. Sleutels, H.V.M. Hamelers, and C.J.N. Buisman. Ammonium recovery and energy production from urine by a microbial fuel cell. *Water Research*, 46(8):2627–2636, 2012.

[97] R. Lal. Soil carbon sequestration to mitigate climate change. *Geoderma*, 123 (1-2):1–22, 2004.

[98] R. Lal, R. F. Follett, B. A. Stewart, and J. M. Kimble. Soil carbon sequestration to mitigate climate change and advance food security. *Soil Science*, 172(12): 943–956, 2007.

[99] E. H. Larsen, N. L. Andersen, A. Møller, A. Petersen, G. K. Mortensen, and J. Petersen. Monitoring the content and intake of trace elements from food in denmark. *Food Additives and Contaminants*, 19(1):33–46, 2002.

[100] T. A. Larsen and J. Lienert. Societal implications of re-engineering the toilet. *Water Intelligence Online March*, 2003.

[101] T. A. Larsen and J. Lienert. Novaquatis final report. NoMix–A new approach to urban water management. Eawag, Dübendorf, 2007.

[102] T. A Larsen, I. Peters, A. Alder, R. Eggen, M. Maurer, and J. Muncke. Re-engineering the toilet for sustainable waste water management. 35(9):192–197, 2001.

[103] T. A. Larsen, A. C. Alder, R. I.L Eggen, M. Maurer, and J. Lienert. Source separation: Will we see a paradigm shift in wastewater handling? *Environmental science & technology*, 43(16):6121–6125, 2009.

[104] T. A. Larsen, K. M. Udert, and J. Lienert, editors. *Source Separation and Decentralization for Wastewater Management.* IWA Publishing, 2013.

[105] K. S. Le Corre, E. Valsami-Jones, P. Hobbs, and S. A. Parsons. Phosphorus recovery from wastewater by struvite crystallization: A review. *Critical Reviews in Environmental Science and Technology*, 39(6):433–477, 2009.

[106] J. C. Leblanc, T. Guérin, L. Noël, G. Calamassi-Tran, J. L. volatier, and P. Verger. Dietary exposure estimates of 18 elements from the 1st french total diet study. *Food Additives and Contaminants*, 22(7):624–641, 2005.

[107] G. Lettinga. Towards feasible and sustainable environmental protection for all. *Aquatic Ecosystem Health & Management*, 11(1):116–124, 2008.

[108] F. Li, K. Wichmann, and R. Otterpohl. Review of the technological approaches for grey water treatment and reuses. *Science of the Total Environment*, 407(11): 3439–3449, 2009.

[109] J. Lienert. High acceptance of source-separating technologies – but... In T. A. Larsen, K. M. Udert, and J. Lienert, editors, *Source Separation and Decentralization for Wastewater Management.* IWA-Publishing, London, UK, 2013.

[110] Y. P. Lin, P. C. Singer, and G. R. Aiken. Inhibition of calcite precipitation by natural organic material: Kinetics, mechanism, and thermodynamics. *Environmental science & technology*, 39(17):6420–6428, 2005.

[111] K. Linderholm, J. E. Mattsson, and A. M. Tillman. Phosphorus flows to and from swedish agriculture and food chain. *Ambio*, 41(8), 2012.

[112] H.W. Lintsen. *Geschiedenis van de techniek in Nederland. De wording van een moderne samenleving 1800-1890. Deel I. Techniek en modernisering. Landbouw en voeding.* Walburg Pers, Zutphen, 1992.

[113] H.W. Lintsen. *Geschiedenis van de techniek in Nederland. De wording van een moderne samenleving 1800-1890. Deel II. Gezondheid en openbare hygiëne. Waterstaat en infrastructuur.* Walburg Pers, Zutphen, 1993.

[114] J. Lu, H. N. Gavala, I. V. Skiadas, Z. Mladenovska, and B. K. Ahring. Improving anaerobic sewage sludge digestion by implementation of a hyper-thermophilic

prehydrolysis step. *Journal of Environmental Management*, 88(4):881–889, 2008.

[115] S. Luostarinen and J. Rintala. Anaerobic on-site treatment of kitchen waste in combination with black water in uasb-septic tanks at low temperatures. *Bioresource technology*, 98(9):1734–1740 0960–8524, 2007.

[116] S. Luostarinen, S. Luste, and M. Sillanpää. Increased biogas production at wastewater treatment plants through co-digestion of sewage sludge with grease trap sludge from a meat processing plant. *Bioresource Technology*, 100(1):79–85, 2009.

[117] J. Ma, M. Van Wambeke, M. Carballa, and W. Verstraete. Improvement of the anaerobic treatment of potato processing wastewater in a UASB reactor by co-digestion with glycerol. *Biotechnology letters*, 30(5):861–867, 2008.

[118] CK Makropoulos, K. Natsis, S. Liu, K. Mittas, and D. Butler. Decision support for sustainable option selection in integrated urban water management. *Environmental Modelling & Software*, 23(12):1448–1460, 2008.

[119] A. Mañas, B. Biscans, and M. Spérandio. Biologically induced phosphorus precipitation in aerobic granular sludge process. *Water research*, 45(12):3776–3786, 2011.

[120] S. Mann. *Biomineralization: principles and concepts in bioinorganic materials chemistry*. Oxford University Press, 2001.

[121] J. Mata-Alvarez, S. Mace, and P. Llabres. Anaerobic digestion of organic solid wastes. an overview of research achievements and perspectives. *Bioresource technology*, 74(1):3–16, 2000.

[122] M. Maurer, P. Schwegler, and T. Larsen. Nutrients in urine: energetic aspects of removal and recovery. *From Nutrient Removal to Recovery*, 48(1):37–46, 2003.

[123] M. Maurer, W. Pronk, and T. A. Larsen. Treatment processes for source-separated urine. *Water Research*, 40(17):3151–3166, 2006.

[124] P. L. McCarty, J. Bae, and J. Kim. Domestic wastewater treatment as a net energy producer–can this be achieved? *Environmental Science & Technology*, 45(17), 2011.

[125] T. Melin, B. Jefferson, D. Bixio, C. Thoeye, W. De Wilde, J. De Koning, J. Van der Graaf, and T. Wintgens. Membrane bioreactor technology for wastewater treatment and reuse. *Desalination*, 187(1-3):271–282, 2006.

[126] B. Meulman, G. Zeeman, and C. J. N. Buisman. Treatment of concentrated black water on pilot scale: options and challenges. In *Sanitation Challenge*, Wageningen, the Netherlands, 2008.

[127] J. K. Miettinen. Absorption and elimination of dietary mercury (hg2+) and methylmercury in man. In *Miller, M.W. & Clarkson, T.W., eds, Mercury, Mercurials and Mercaptans*, pages 233–243. Charles C Thomas, Springfield IL, USA, 1973.

[128] C. Minoia, E. Sabbioni, P. 1 Apostoli, R. Pietra, L. Pozzoli, M. Gallorini, G. Nicolaou, L. Alessio, and E. Capodaglio. Trace element reference values in tissues from inhabitants of the european community i. a study of 46 elements in urine, blood and serum of italian subjects. *Science of the total environment*, 95:89–105, 1990.

[129] Y. Miron, G. Zeeman, J. B. Van Lier, and G. Lettinga. The role of sludge retention time in the hydrolysis and acidification of lipids, carbohydrates and proteins during digestion of primary sludge in cstr systems. *Water Research*, 34(5):1705–1713 0043–1354, 2000.

[130] K. Moriyama, T. Mori, H. Arayashiki, H. Saito, and M. Chino. The amount of heavy metals derived from domestic wastewater. *Water Science & Technology*, 21:1913–1916, 1989.

[131] J. A. Müller, L. Günther, T. Dockhorn, N. Dichtl, L. C. Phan, I. Urban, D. Weichgrebe, K. H. Rosenwinkel, and N. Bayerle. *Nutrient Recycling from Sewage Sludge using the Seaborne Process*, pages 629–633. GMSC, 2007.

[132] T. H. Muster, G. B. Douglas, N. Sherman, A. Seeber, N. Wright, and Y. Güzükara. Towards effective phosphorus recycling from wastewater: Quantity and quality. *Chemosphere*, 91(5):676–684, 2013.

[133] F. A. Nicholson, S. R. Smith, B. J. Alloway, C. Carlton-Smith, and B. J. Chambers. An inventory of heavy metals inputs to agricultural soils in england and wales. *Science of The Total Environment*, 311(1-3):205–219, 2003.

[134] C. Niwagaba, M. Nalubega, B. Vinnerås, C. Sundberg, and H. Jönsson. Bench-scale composting of source-separated human faeces for sanitation. *Waste management*, 29(2), 2009.

[135] G. Nziguheba and E. Smolders. Inputs of trace elements in agricultural soils via phosphate fertilizers in european countries. *Science of The Total Environment*, 390(1):53–57, 2008.

[136] E. G. Offenbacher, H. Spencer, H. J. Dowling, and F. X. Pi-Sunyer. Metabolic chromium balances in men. *The American journal of clinical nutrition*, 44(1): 77–82, 1986.

[137] OLI systems. Simulation software for electrolyte chemistry, 2012. Accessed on: 22 August 2013. URL `http://www.olisystems.com/`.

[138] Ostara. Crystal green: All ways green (OSTARA), 2009. Accessed on: 22 August 2013. URL `http://www.crystalgreen.com/`.

[139] M. Ottaviani, A. Santarsiero, and S. De Fulvio. Heavy metals in sewage sludge utilized in agriculture. *Ann. Isl. Super. Sanita*, 25(3):525–530, 1989.

[140] R. Otterpohl, M. Grottker, and J. Lange. Sustainable water and waste management in urban areas. *Water Science & Technology*, 35(9):121–133, 1997.

[141] R. Otterpohl, U. Braun, and M. Oldenburg. Innovative technologicies for decentralized water-, wastewater and biowaste management in urban and peri-urban areas. *Water Science & Technology*, 48(11-12):23–32, 2003.

[142] L. M. Ottosen, G. M. Kirkelund, and P. E. Jensen. Extracting phosphorous from incinerated sewage sludge ash rich in iron or aluminum. *Chemosphere*, 91 (7):963–969, 2013.

[143] H. Palmquist and J. Hanæus. Hazardous substances in separately collected grey- and blackwater from ordinary swedish households. *Science of The Total Environment*, 348(1-3):151–163, 2005.

[144] J. Pinnekamp and D. Montag. *Phosphorrückgewinnung aus Überschussschlamm*. 2005.

[145] L. J. Plant and W. A. House. Precipitation of calcite in the presence of inorganic phosphate. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, 203(1):143–153, 2002.

[146] R. Prasad, A. Britton, B. Balzer, and G. Schafran. Nutrient recovery by struvite crystallization process: virginia experience. *Proceedings of the Water Environment Federation*, 2007(19), 2007.

[147] P. Prinzhorn. Phosphordünger aus klärschlammaschen mit thermischer schwermetallentfrachtung. In *75. Darmstädter Seminar–Rückgewinnung von Phosphor aus Abwasser und Klärschlamm, Schriftenreihe WAR, Nr. 167*, Darmstadt, Germany, 2005.

[148] I. Rehman and W. Bonfield. Characterization of hydroxyapatite and carbonated apatite by photo acoustic FTIR spectroscopy. *Journal of Materials Science: Materials in Medicine*, 8(1):1–4, 1997.

[149] C. Remy and A. Ruhland. Ecological assessment of alternative sanitation concepts with life cycle assessment. *Berlin, Germany: Technical University Berlin*, 55, 2006.

[150] R. Renner. Sewage sludge, pros & cons. *Environmental science & technology*, 34(19):430A–435A, 2000.

[151] RIONED foundation. Urban drainage statistics 2009-2010, 2013. Accessed on: 3 February 2013. URL `http://www.riool.net/`.

[152] M. Rose, M. Baxter, N. Brereton, and C. Baskaran. Dietary exposure to metals and other elements in the 2006 UK total diet study and some trends over the last 30 years. *Food Additives & Contaminants: Part A*, 27(10):1380–1404, 2010.

[153] A. Rosmarin. The precarious geopolitics of phosphorus, down to earth. *Science and environment Fortnightly*, 30:27–31, 2004.

[154] T. Rossman. Arsenic. In *Environmental and occupational medicine*, pages 1006–1017. Lippincott Williams & Wilkins, Rom W, Markowitz S (eds), Hagerstown, MD, 4th edition, 2007.

[155] K. L. Rule, S. D. W. Comber, D. Ross, A. Thornton, C. K. Makropoulos, and R. Rautiu. Diffuse sources of heavy metals entering an urban wastewater catchment. *Chemosphere*, 63(1):64–72, March 2006.

[156] H. Sälzer, T. Ochs, and Rüster C. Research project sanitaryrecycling eschborn (SANIRESCH) factsheet - project component: Sanitary and in-house installations. Deutsche Gesellschaft für Internationale Zusammenarbeit GmbH, 2012.

[157] C. H. Saueia, B. P. Mazzilli, and D. I. T. Fávaro. Natural radioactivity in phosphate rock, phosphogypsum and phosphate fertilizers in brazil. *Journal of Radioanalytical and Nuclear Chemistry*, 264(2), 2005.

[158] C. Schaum. *Verfahren für eine zukünftige Klärschlammbehandlung – Klärschlammkonditionierung und Rückgewinnung von Phosphor aus Klärschlammasche*. PhD thesis, Darmstadt, Germany, 2007.

[159] W. J. Schipper, A. Klapwijk, B. Potjer, W. H. Rulkens, B. G. Temmink, F.D. G. Kiestra, and A.C. M. Lijmbach. Phosphate recycling in the phosphorus industry. *Environmental Technology*, 22(11):1337–1345, 2001.

[160] C. Schönning and T. A. Stenström. *Guidelines on the safe use of urine and faeces in ecological sanitation systems*. EcoSanRes Publication Series, Report 2004-1, Stockholm Environment Insitute, Stockholm, Sweden, 2004.

[161] N. L. Schouw, S. Danteravanich, H. Mosbaek, and J. C. Tjell. Composition of human excreta - a case study from southern thailand. *the Science of the Total Environment*, 286:155–166, 2002.

[162] P. Schramel, I. Wendler, and J. Angerer. The determination of metals (antimony, bismuth, lead, cadmium, mercury, palladium, platinum, tellurium, thallium, tin and tungsten) in urine samples by inductively coupled plasma-mass spectrometry. *International archives of occupational and environmental health*, 69(3):219–223, 1997.

[163] J. J. Schröder, D. Cordell, A. L. Smit, and A. Rosemarin. Sustainable use of phosphorus. *Wageningen: Plant Research International*, 2010.

[164] R. D. Schuiling and A. Andrade. Recovery of struvite from calf manure. *Environmental Technology*, 20(7):765–768, 1999.

[165] University of the West of England Science Communication Unit. *Science for Environment Policy In-depth Report: Sustainable Phosphorus Use.* Report produced for the European Commission DG Environment, 2013.

[166] M. M. Seckler, O. S. L. Bruinsma, and G. M. Van Rosmalen. Phosphate removal in a fluidized bed—I. identification of physical processes. *Water Research*, 30 (7):1585–1588, 1996.

[167] L. Shu, P. Schneider, V. Jegatheesan, and J. Johnson. An economic evaluation of phosphorus recovery as struvite from digester supernatant. *Bioresource Technology*, 97(17), 2006.

[168] R. P. Singh and M. Agrawal. Potential benefits and risks of land application of sewage sludge. *Waste Management*, 28(2):347–358, 2008.

[169] Y. Song, P. G Weidler, U. Berg, R. Nüesch, and D. Donnert. Calcite-seeded crystallization of calcium phosphate for phosphorus recovery. *Chemosphere*, 63 (2):236–243, 2006.

[170] L. Sörme and R. Lagerkvist. Sources of heavy metals in urban wastewater in stockholm. *Science of the Total Environment*, 298(1):131–145, 2002.

[171] P. Sosnowski, A. Wieczorek, and S. Ledakowicz. Anaerobic co-digestion of sewage sludge and organic fraction of municipal solid wastes. *Advances in Environmental Research*, 7(3):609–616, 2003.

[172] J. Spångberg. *Recycling plant nutrients from waste and by-products.* PhD thesis, Swedish University of Agricultural Sciences, 2014.

[173] T. S. Srikumar, G. K. Johansson, P. A. Öckerman, J. Å. Gustafsson, and B. Åkesson. Trace element status in healthy subjects switching from a mixed to a lactovegetarian diet for 12 mo1-3. *The American Journal of Clinical Nutrition*, 55(4):885–890, 1992.

[174] T. A. Stenström. Hygiene, a major challenge for source separation and decentralization. In T. A. Larsen, K. M. Udert, and J. Lienert, editors, *Source Separation and Decentralization for Wastewater Management.* IWA Publishing, London, UK, 2013.

[175] E. Storr-Hansen and S. C. Rastogi. Polychlorinated biphenyls and heavy metal levels in recycled paper for household use. *Bulletin of environmental contamination and toxicology*, 40(3):451–456, 1988.

[176] SUSAN. Sustainable and Safe Re-use of Municipal Sewage Sludge for Nutrient Recovery, 2008. Accessed on: 1 May 2014. URL `http://www.susan.bam.de`.

[177] K. Suzuki, Y. Tanaka, K. Kuroda, D. Hanajima, Y. Fukumoto, T. Yasuda, and M. Waki. Removal and recovery of phosphorous from swine wastewater by demonstration crystallization reactor and struvite accumulation device. *Bioresource Technology*, 98(8):1573–1578, 2007.

[178] G. Tamasi and R. Cini. Heavy metals in drinking waters from mount amiata (tuscany, italy). possible risks from arsenic for public health in the province of siena. *Science of The Total Environment*, 327(1-3):41–51, 2004.

[179] H. Tapiero and K. D. Tew. Trace elements in human physiology and pathology: zinc and metallothioneins. *Biomedicine & Pharmacotherapy*, 57(9):399–411, 2003.

[180] H. Tapiero, D.M. Townsend, and K.D. Tew. Trace elements in human physiology and pathology. copper. *Biomedicine & Pharmacotherapy*, 57(9):386–398, 2003.

[181] G. Tchobanoglous, F. L. Burton, and H. D. Stensel. *Wastewater Engineering: treatment and reuse.* McGraw-Hill series in civil and environmental engineering (M. E. Inc., Ed.). McGraw-Hill, New York, 4th edition, 2004.

[182] T.A. Ternes and A. Joss. *Human pharmaceuticals, hormones and fragrances: the challenge of micropollutants in urban water management.* International Water Association, 2006.

[183] T. Tervahauta, T. Hoang, L. Hernández Leal, G. Zeeman, and C. J. N. Buisman. Prospects of source-separation-based sanitation concepts: A model-based study. *Water*, 5(3):1006–1035, 2013.

[184] T. Tervahauta, S. Rani, L. Hernández Leal, C. J. N. Buisman, and G. Zeeman. Black water sludge reuse in agriculture: Are heavy metals a problem? *Journal of Hazardous Materials*, 274:229–236, 2014.

[185] T. Tervahauta, R. D. van der Weijden, R. L. Flemming, L. Hernández Leal, G. Zeeman, and C. J. N. J.N. Buisman. Calcium phosphate granulation in anaerobic treatment of black water: A new approach to phosphorus recovery. *Water Research*, 48:632–642, 2014.

[186] C. Thibodeau, F. Monette, M. Glaus, and C. B. Laflamme. Economic viability and critical influencing factors assessment of black water and grey water source-separation sanitation system. *Water Science & Technology*, 64(12):2417–2424, 2011.

[187] P. Tidåker, B. Mattsson, and H. Jönsson. Environmental impact of wheat production using human urine and mineral fertilisers – a scenario study. *Journal of Cleaner Production*, 15(1):52–62, 2007.

[188] E. Tilley, J. Atwater, and D. Mavinic. Recovery of struvite from stored human urine. *Environmental technology*, 29(7), 2008.

[189] G. Turconi, C. Minoia, A. Ronchi, and C. Roggi. Dietary exposure estimates of twenty-one trace elements from a total diet study carried out in pavia, northern italy. *British Journal of Nutrition*, 101(08):1200, 2008.

[190] UCPTE. Yearly report 1993, 1994.

[191] Y. Ueno. *Full scale struvite recovery in Japan.* Phosphorus in Environmental Technology. IWA-Publishing, London, UK, 2004.

[192] Y. Ueno and M. Fujii. Three years experience of operating and selling recovered struvite from full-scale plant. *Environmental Technology*, 22(11):1373–1381, November 2001.

[193] USGS. *Fertilizers - Sustaining global food supplies.* US Department of the Interior, US Geological Survey, Reston, USA, 1999.

[194] M. Vahter, M. Berglund, B. Lind, L. Jorhem, S. Slorach, and L. Friberg. Personal monitoring of lead and cadmium exposure–a swedish study with special reference to methodological aspects. *Scandinavian Journal of Work, Environment & Health*, 17(1):65–74, 1991.

[195] M. Vahter, M. Berglund, S. Slorach, L. Friberg, M. Sarić, X. Zheng, and M. Fujita. Methods for integrated exposure monitoring of lead and cadmium. *Environmental research*, 56(1):78–89, 1991.

[196] M. Vahter, G. Johansson, A. Åkesson, and B. Rahnster. Faecal elimination of lead and cadmium in subjects on a mixed and a lactovegetarian diet. *Food and chemical toxicology*, 30(4):281–287, 1992.

[197] C. L. van Beek. Food security in a fast-changing world: a new approach to improving soil fertility, 2013.

[198] I. van Beuzekom, S. Dekker, and M. Teunissen. Duurzame afvalwaterbehandeling geerpark heusden. Grontmij, 2010.

[199] J. C. L. van Buuren. *Sanitation choice involving stakeholders.* PhD thesis, University of Wageningen, the Netherlands, 2010.

[200] M. van der Wijst and A. P. Groot-Marcus. *Huishoudelijk afvalwater: Berekening van de zuurstofvraag.* Wageningen Landbouuniversiteit, 1998.

[201] K. van Dijk, J. Sanders, P. Groot Koerkamp, B. Smit, P. Ehlert, O. Schoumans, G. Zeeman, and O. Oenema. *Exploring options for a transition towards sustainable use of phosphorus: Food chain studies for the Netherlands and EU.* Alterra, Wageningen UR, 2012.

[202] H. J. C. van Dooren, M. C. Hanegraaf, and K. Blanken. Emission and compost quality of on farm composted dairy slurry. PraktijkRapport rundvee 68, animal sciences group, 2005.

[203] M. Vargová, O. Ondrašovičová, N. Sasáková, M. Ondrašovič, K. Čulenová, and S. Šmirjáková. Heavy metals in sewage sludge and pig slurry solids and the health and environmental risk associated with their application to agricultural soil. *Folia Veterinaria*, 49(3):28–30, 2005.

[204] A. Veeken and B. Hamelers. Sources of cd, cu, pb and zn in biowaste. *Science of the Total Environment*, 300(1):87–98, 2002.

[205] Willy Verstraete and Siegfried E. Vlaeminck. ZeroWasteWater: short-cycling of wastewater resources for sustainable cities of the future. *International Journal of Sustainable Development & World Ecology*, 18:253–264, 2011.

[206] B. Vinnerås, H. Palmquist, P. Balmér, and H. Jönsson. The characteristics of household wastewater and biodegradable solid waste—A proposal for new swedish design values. *Urban Water Journal*, 3(1):3–11, 2006.

[207] B. Vinnerås, A. Nordin, C. Niwagaba, and K. Nyberg. Inactivation of bacteria and viruses in human urine depending on temperature and dilution rate. *Water Research*, 42(15):4067–4074, 2008.

[208] Björn Vinnerås. *Possibilities for sustainable nutrient recycling by faecal separation combined with urine.* PhD thesis, Swedish University of Agricultural Sciences, Uppsala, Sweden, 2002.

[209] Björn Vinnerås. Comparison of composting, storage and urea treatment for sanitising of faecal matter and manure. *Bioresource Technology*, 98(17), 2007.

[210] Vitens. Drinking water quality in friesland (NL). Vitens, 2012. Accessed on: 22 August 2013. URL `http://www.vitens.nl/`.

[211] Vitens. Drinking water quality in friesland (NL). Vitens, 2013. Accessed on: 8 September 2013. URL `http://www.vitens.nl/`.

[212] S. E. Vlaeminck, A. Terada, B. F. Smets, D. Linden, N. Boon, W. Verstraete, and M. Carballa. Nitrogen removal from digested black water by one-stage partial nitritation and anammox. *Environmental Science & Technology*, 43 (13):5035–5041, 2009.

[213] WHO. *Arsenic (International Programme on Chemical Safety, Environmental Health Criteria, 18).* World Health Organization, Geneva, Switzerland, 1981.

[214] WHO. *Cadmium (International Programme on Chemical Safety, Environmental Health Criteria, 134).* World Health Organization, Geneva, Switzerland, 1992.

[215] WHO. *Guidelines for the Safe Use of Wastewater, Excreta and Greywater, Volume 3: Wastewater and Excreta Use in Aquaculture.* World Health Organization, 2006.

[216] WHO. *Guidelines for the safe reuse of wastewater, excreta and greywater, vol. 1–4.* World Health Organization, Geneva, Switzerland., 2006.

[217] J. A. Wilsenach and M. C. M. van Loosdrecht. Integration of processes to treat wastewater and source-separated urine. *Journal of Environmental Engineering*, 132(3):331–341, 2006.

[218] J. A. Wilsenach, M. Maurer, T. A. Larsen, and M. C. M. van Loosdrecht. From waste treatment to integrated resource management. *Water Science & Technology*, 48(1):1–9, 2003.

[219] J. A. Wilsenach, C. A. H. Schuurbiers, and M. C. M. Van Loosdrecht. Phosphate and potassium recovery from source separated urine through struvite precipitation. *Water research*, 41(2), 2007.

[220] M. Winker, B. Vinnerås, A. Muskolus, U. Arnold, and J. Clemens. Fertiliser products from new sanitation systems: Their potential values and risks. *Bioresource technology*, 100(18):4090–4096, 2009.

[221] M. Winker, J. Clemens, M. Reich, H. Gulyas, and R. Otterpohl. Ryegrass uptake of carbamazepine and ibuprofen applied by urine fertilization. *Science of The Total Environment*, 408(8):1902–1908, 2010.

[222] E. Wittig. Erste betriebserfahrungen der klärschlammaufbereitungsanlage nach dem seaborne- verfahren/gifhorner-verfahren. In *30. Erfahrungsaustausch der Lehrerinnen und Lehrer norddeutscher Kläranlagen-and Kanalnachbarschaften in Lüneburg, Germany*, 2007.

[223] G. Ysart, P. Miller, H. Crews, P. Robb, M. Baxter, C. De L'Argy, S. Lofthouse, C. Sargent, and N. Harrison. Dietary exposure estimates of 30 elements from the UK total diet study. *Food Additives and Contaminants*, 16(9):391–403, 1999.

[224] G. Zeeman. *New sanitation: bridging cities and agriculture.* Wageningen University, 2012.

[225] G. Zeeman and K. Kujawa-Roeleveld. Resource recovery from source separated domestic waste(water) streams; full scale results. *Water Science & Technology*, 64(10):1987, 2011.

[226] G. Zeeman and G. Lettinga. The role of anaerobic digestion of domestic sewage in closing the water and nutrient cycle at community level. *Water Science & Technology*, 39(5):187–194, 1999.

[227] G. Zeeman, K. Kujawa, T. de Mes, L. Hernandez, M. de Graaff, L. Abu-Ghunmi, A. Mels, B. Meulman, H. Temmink, C. J. N. Buisman, J. van Lier, and G. Lettinga. Anaerobic treatment as a core technology for energy, nutrients and water recovery from source-separated domestic waste(water). *Water Science and Technology*, 57(8):1207, 2008.

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Taina Tervahauta
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About the author

Taina Tervahauta was born on 15 March 1984 in Turku, Finland. In 2003 she started her study of water and waste management engineering at Tampere University of Technology in the department of Environmental and Energy Technology. She finished her Master of Science degree by completing a thesis on removal of antiscalants from membrane concentrates using adsorption technology at Wetsus, centre of excellence for sustainable water technology in the Netherlands in 2010. After gaining experience in drinking water related research, she found her true passion in resource recovery from source-separated domestic wastewater. She started research on phosphate and organic fertilizer recovery from black water as a PhD student at the Sub-department of Environmental Technology in Wageningen University, but was stationed at Wetsus in Leeuwarden.

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