



National Institute for Public Health
and the Environment
Ministry of Health, Welfare and Sport

Recycling of waste streams containing human and veterinary pharmaceuticals

An overview of technological developments and possible
consequences for pharmaceutical releases into the
environment

RIVM Letter report 2015-0174

J.G.M. Derksen | C.T.A. Moermond | C.W.M. Bodar



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Colophon

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Recycling van afvalstromen met resten van (dier)geneesmiddelen

Afvalwater en mest bevatten waardevolle grondstoffen waaraan energie kan worden onttrokken. Ook kunnen er nieuwe producten van worden gemaakt, zoals bioplastics, bouwmaterialen en papier. De laatste tijd worden veel nieuwe technieken ontwikkeld die de recycling van afvalwater en mest mogelijk maken. Het is echter vaak nog niet duidelijk wat de invloed van deze recycling is op de mate waarin restanten van (dier)geneesmiddelen in het milieu terecht komen. Dit blijkt uit onderzoek van het RIVM.

Het RIVM inventariseerde hiervoor de innovaties bij de afvalwater- en mestverwerking en onderzocht hoe deze technieken van invloed zijn op de emissie van (dier)geneesmiddelen naar het milieu. Meer onderzoek is nodig om te bepalen of, en zo ja in hoeverre, dit gebeurt. In de meeste gevallen ontbreken hiervoor nog de benodigde meetgegevens.

Milieu-emissies van (dier)geneesmiddelen

Diverse nieuwe zuiveringstechnieken voor afvalwater zorgen er naar verwachting voor dat er minder restanten van (dier)geneesmiddelen in het milieu komen. Maar het is bijvoorbeeld nog niet duidelijk of de emissie van (dier)geneesmiddelen naar het milieu verandert bij mestverwerking, of bij de (co)vergisting van slib en/of mest. (Co)vergisting levert biogas op, maar mogelijk blijven hoge concentraties (dier)geneesmiddelen over in het residu dat als mest wordt aangewend. Ook is onduidelijk hoeveel (dier)geneesmiddelen achterblijven in grondstoffen die ontstaan uit de afvalwaterzuivering, zoals bij de meststof struviet.

Aanbevelingen voor beleid

De milieurisicobeoordeling bij de Europese registratie van (dier)geneesmiddelen houdt nu nog geen rekening met de nieuwe verwerkingstechnieken voor afvalwater en mest. Het RIVM vindt het raadzaam om op EU-niveau de toekomstige technologische ontwikkelingen nauwlettend te volgen. Vanwege de bovengenoemde onduidelijkheden lijkt het echter nog te vroeg om de algemene Europese beoordelingsmethodologie te wijzigen. Verder pleit het RIVM ervoor dat er meer informatie beschikbaar komt over de omvang van de emissies van (dier)geneesmiddelen via de verschillende recyclingroutes. Dergelijke gegevens zijn ook belangrijk bij het afwegen van veiligheids- en duurzaamheidsaspecten in een circulaire economie.

Kernwoorden: geneesmiddelen, afvalwater, mest, hergebruik, circulaire economie

Abstract

Recycling of waste streams containing human and veterinary pharmaceuticals

Wastewater and manure contain valuable raw materials to produce either energy sources or products, such as bioplastics, construction materials and papier. At present many new technologies are being developed to recycle wastewater and manure. The impact of this recycling or reuse on the environmental release of residues of veterinary or human drugs is mostly still unknown.

RIVM investigated the current innovations in processing wastewater and manure and studied how these technologies may affect the emissions of (veterinary) pharmaceuticals to the environment. More research is needed to determine if emissions are being influenced, and, if yes, to what extent. In most cases sound monitoring data were found to be lacking.

Environmental emissions of (veterinary) pharmaceuticals

Various new wastewater sanitation techniques will most probably reduce the environmental releases of (veterinary) pharmaceuticals. It is not clear, however, if these emissions will change at the processing of manure, or at co-digestion of sludge and/or manure. Co-digestion produces biogas, but high concentrations of (veterinary) drugs may remain in the residue that is being applied as fertilizer. The same may hold for resources from wastewater recycling, such as the fertilizer struvite.

Policy recommendations

The environmental risk assessment at the European registration of (veterinary) pharmaceuticals does not yet take into account these new process technologies for wastewater and manure. RIVM advises to accurately follow the technological developments. However, it would be premature to adapt the generic EU risk assessment methodology now already, because of the abovementioned uncertainties.

RIVM further endorses that more information becomes available on the scale of the emissions of veterinary and human drugs from the various recycling pathways. Such data are also important when weighing safety versus sustainability aspects in circular economy developments.

Keywords: pharmaceuticals, waste water, manure, re-use, circular economy

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Summary

The transition towards a circular economy, *i.e.* moving 'from waste to resource', is a very promising development from both economic, environmental health and sustainability perspective. However, new waste recycling or reuse routes may also encompass several points of attention. Waste streams containing micropollutants may be withdrawn from conventional treatment systems and may thus enter the environment in different amounts than before or within other emission routes than before.

The scope of this report was to give a brief overview of the new technological developments in waste treatment and recycling, focussing on waste containing human and veterinary pharmaceuticals. An identification was given on the 'status' of the technology, *i.e.* a distinction is made between the exploratory (lab scale or pilot plant) or operational stage. Additionally, an assessment was made on the potential influence of the processes on the emission of (veterinary) pharmaceuticals into the environment.

When analysing the new technologies in many cases quantitative data on the actual occurrence or mass balances of human and veterinary pharmaceuticals was found to be lacking. This implies that judgements on possible changes of environmental loads could 'only' be qualitatively scored as potential increase, potential decrease or no change.

Initiatives such as post treatment of sewage treatment plant (STP) effluent or separate treatment of hospital wastewater will undoubtedly lower the emission of pharmaceuticals to the environment. However, the following technologies could be of concern because of either possible increases of pharmaceutical emissions or unfavourable changes in their exposure routes: products recovered from the STP, digestion of sewage sludge, use of concentrates to replace chemical fertilizer, co-digestion or composting of manure, and treatment of manure together with wastewater at the STP.

The current study further indicated that several resource recovery and sanitation technologies are still in an experimental stage. Once brought forward to the operational stage, these new technologies will influence, either positively or negatively, the environmental load and potential risk of human and veterinary pharmaceuticals. It is concluded, however, that it seems too early to adapt now already the EU Environmental Risk Assessment (ERA) scheme at the registration of (veterinary) pharmaceuticals. Nevertheless RIVM recommends to further address the topic at EU level. It should be emphasised that both new sanitation concepts and circular economy applications will eventually influence the methodology for assessing environmental risks of (veterinary) pharmaceuticals.

RIVM also endorses the need for more quantitative information on the emissions and fate of (veterinary) pharmaceuticals in the various waste treatments and recycling pathways. Such data are also important when

weighing safety versus sustainability aspects in circular economy developments.

The waste streams that were considered in the report may not only contain (veterinary) pharmaceuticals, but also other micropollutants, such as biocides, cleaning agents and microplastics. Many remarks that were made on (veterinary) pharmaceuticals may therefore also apply to these substances.

1 Introduction

1.1 General

The Dutch government aims to promote sustainable use and reuse of natural resources. The recovery of raw materials from waste or communal wastewater can help prevent resource depletion, reduce dependence on supplies, and save energy. The ambition is to promote 1) a circular economy, *i.e.* a transition 'from waste to resource', 2) sustainable energy and 3) a biobased economy, *i.e.* use of renewable biomass instead of oil based products (MinEZ, 2013).

Waste streams and/or wastewater may contain valuable resources for energy and products, but may also contain residues of human or veterinary pharmaceuticals, including hormones, pathogens and other contaminants. As this report is written within the framework of 'Policy advice on pharmaceuticals and environment', the main focus of this report is on pharmaceuticals. Furthermore, the target is on communal wastewater containing human excreta and agricultural waste streams containing animal excreta. Whenever the term wastewater is used, the communal wastewater (sewage) is meant (versus industrial wastewater).

When the way in which waste (water) streams are treated changes because of reuse or else, the emission routes and amounts of pharmaceuticals to the environment may also change. In some cases this could result in a reduction of the environmental load, while in other cases the pharmaceutical release to the environment may increase.

Political attention for pharmaceuticals in the environment has increased, both at national and European levels. The presence of pharmaceuticals and their possible effects on the aquatic ecosystem has led to the placement of a number of pharmaceuticals on the so-called 'watch list' under the Water Framework Directive. In 2015, the EU Commission has to come up with a strategy on how to deal with pharmaceuticals in the environment. For the EU marketing authorization of human and veterinary pharmaceuticals, an environmental risk assessment (ERA) has to be performed. The environmental risk of pharmaceuticals is assessed using standardized models, with a number of possible, conventional, release routes of these substances into the environment. When the routes and rates of pharmaceutical release change due to current or future reuse processes, this may have to be taken into account in the models used to assess the environmental risk.

In this report an inventory will be made of new routes of treatment and use of wastewater and manure containing pharmaceuticals. The inventory will focus on the potential changes in release of pharmaceuticals to the environment.

First of all - as a starting point - the current entry routes of pharmaceuticals into the environment, and the legislative framework will be discussed, for both human pharmaceuticals (paragraph 1.2) and

veterinary pharmaceuticals (paragraph 1.3). In paragraph 1.4 the aim and scope of the report will be further discussed.

1.2 Human pharmaceuticals in waste

1.2.1 Entry into the environment

Due to the use of pharmaceuticals, human urine and faeces contain residues of these pharmaceuticals. These residues may consist of the parent compound (the compound which exerts the effect in the body), but may also consist of one or more metabolites of this parent compound. Most of the pharmaceutical residues are excreted via urine, a smaller part is excreted with the faeces. With urine and faeces, pharmaceuticals enter the sewage system and subsequently end up in sewage treatment facilities. Conventionally, these facilities were designed to remove nutrients and suspended solids from the sewage, and not specifically aimed to remove micropollutants, like pharmaceuticals. Despite biodegradation and sorption to sewage sludge, pharmaceutical residues are only partly removed by sewage treatment facilities and therefore residues may end up in surface water. Sewage sludge is usually incinerated in the Netherlands, which effectively removes all pharmaceuticals (unlike some other EU countries where the sewage sludge is still deposited in landfills or used as fertilizer).

Because pharmaceuticals are very diverse in their physico-chemical properties, no general statements can be made on the route of entry into the sewage system (*i.e.* urine/faeces; parent/metabolites), nor on the treatment success in sewage treatment plants.

Besides entering the environment through metabolic excretion, human pharmaceuticals may also enter the environment when leftover medication is thrown in the waste bin or flushed away through the toilet.

1.2.2 Legislative framework

In the legal framework for the marketing authorisation for human pharmaceuticals (Directive 2001/83/EC, as amended) it is posed that 'an indication of any potential risks presented by the medicinal product for the environment' should be part of the dossier, and 'specific arrangement to limit it [environmental risks] shall be envisaged'.

Within the environmental risk analysis, the sole route of entry of pharmaceuticals into the environment is through the sewage treatment plant (EMA, 2006). Depending on the physico-chemical properties of the compound, it is assumed that it may end up in surface water, sediment, groundwater and soil (via sewage sludge). The routes that are taken into account in the environmental risk assessment for marketing authorisation for human pharmaceuticals are illustrated in Figure 1.

Specific frameworks aim to limit the effects of micropollutants, including pharmaceuticals, on the environment (*e.g.*, Water Framework Directive, Groundwater Directive, Soil Directive). However, mutual coordination between these legislations and the Pharmaceutical Directive is lacking.

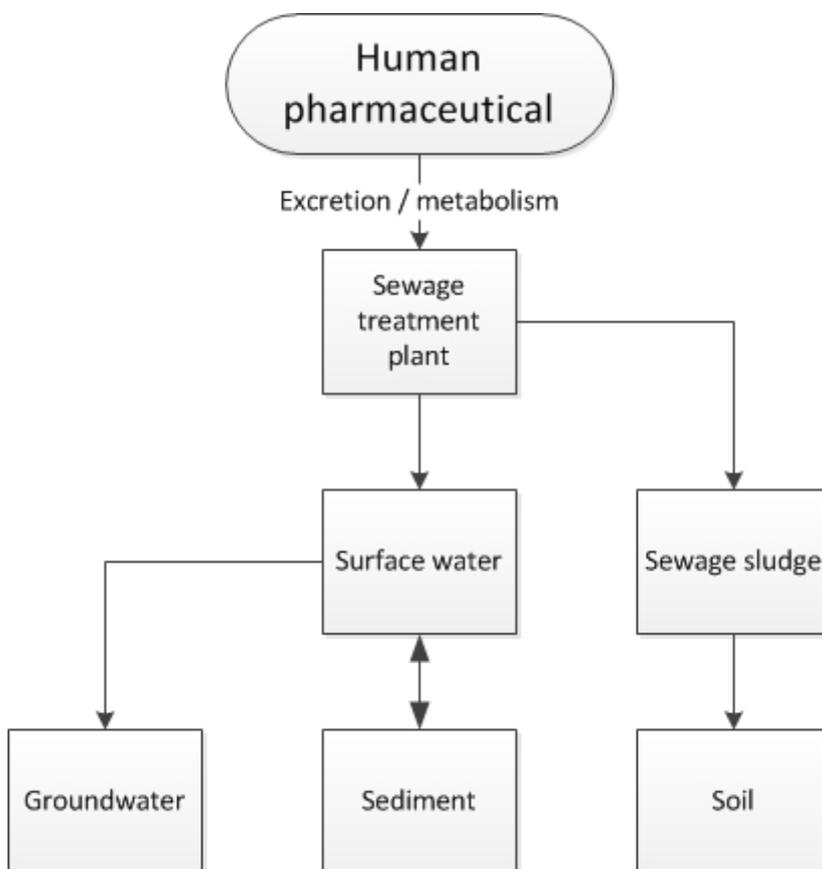


Figure 1 Routes that are taken into account in the environmental risk assessment for marketing authorisation for human pharmaceuticals (for guidance see EMA, 2006). Please note that the sewage sludge-soil route is not relevant for the Netherlands.

1.3 Veterinary pharmaceuticals in waste

1.3.1 Entry into the environment

Where for human pharmaceuticals the route of entry into the environment is relatively straight-forward, veterinary pharmaceuticals are used in many different ways and subsequently also enter the environment in many ways. Within the environmental risk assessment for the marketing authorisation process, a number of routes are taken into account (EMA, 2005). These routes concern mainly food-producing animals, like cows, pigs, horses, sheep, goats, poultry and fish farms. It is assumed that the terrestrial environment is exposed to pharmaceuticals via (1) direct excretion by grazing animals; (2) loss from animals treated on the skin; (3) application of manure, slurry or sludge as fertiliser. The aquatic environment is exposed via (1) leaching, run-off and drainage from manured land; (2) direct spillage and/or food spillage; (3) direct excretion into water by pasture animals; (4) direct application in water for aquaculture; (5) direct discharge of wastewater into surface water (indoor aquaculture); (6) release to sewage treatment plants (indoor aquaculture). The most important routes in the environmental risk assessment for the marketing authorisation of

veterinary pharmaceuticals to the environment are illustrated in Figure 2.

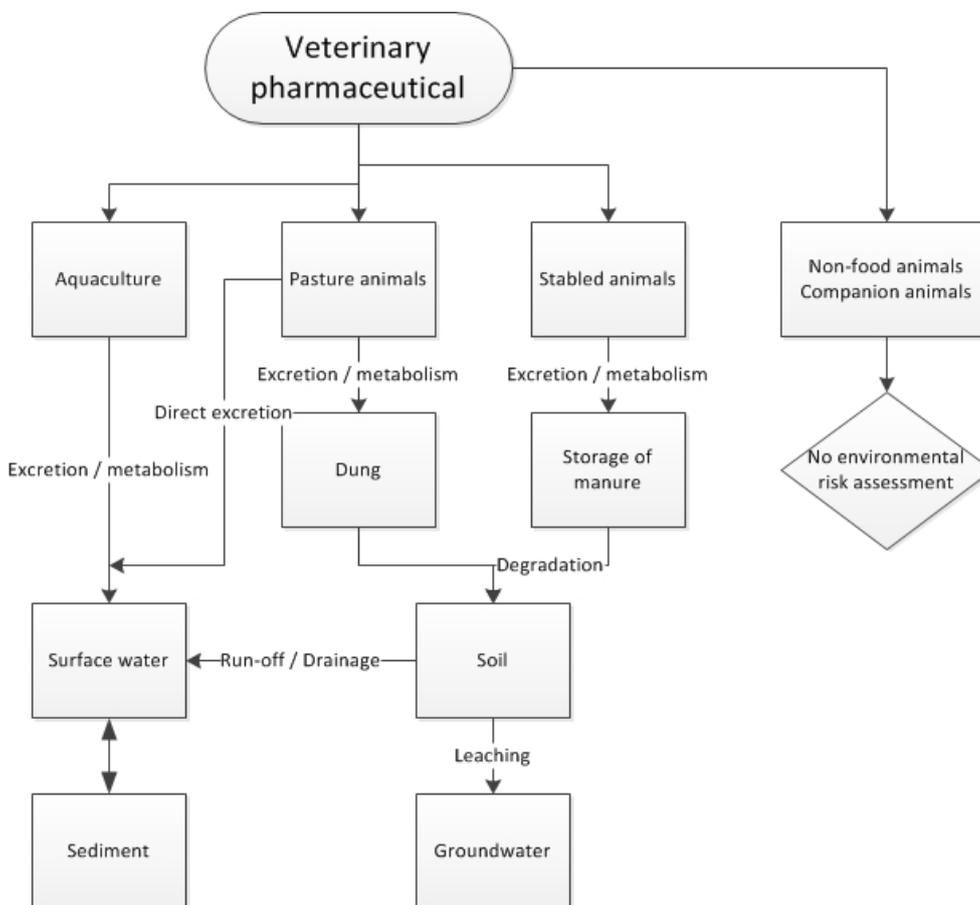


Figure 2 Routes that are taken into account in the environmental risk assessment for marketing authorisation for veterinary pharmaceuticals (for guidance: see EMA, 2005).

Depending on the route of entry into the environment, the veterinary pharmaceutical may end up in soil, groundwater, surface water and/or sediment. How much of the compound ends up in which compartment, depends on the physico-chemical parameters of the compound, the use scenario of the product and characteristics of the environmental compartments exposed. Within the environmental risk assessment, a model calculation is used to estimate this.

Veterinary pharmaceuticals that are used for domestic or pet animals (from donkeys to fish to dogs to rabbits to snakes) can enter the environment in many different ways, depending on the pet. These are not subject to an environmental risk assessment in the marketing authorisation process of these pharmaceuticals and, because of this, their diversity, and because they are mainly disposed of via the waste bin, are not taken into account in this report.

1.3.2 *Legislative framework*

Directive 2001/82/EC as amended requires an environmental risk assessment to be performed for the marketing authorisation of veterinary pharmaceuticals. Like for human pharmaceuticals, a mutual coordination between the veterinary pharmaceutical framework and other EU legal frameworks on environmental quality protection is lacking.

1.4 **Aim and scope of this report**

Currently, the amount of initiatives regarding the reuse of waste as source for energy and other products is increasing. Changes in the way human and veterinary waste is treated and/or used may cause different entry paths of human and veterinary pharmaceuticals in the environment, which are not yet taken into account in the legislative frameworks. The scope of this report is to give an overview of the new technological developments in waste treatment and re-use, focussing on waste containing pharmaceuticals.

A potential change of the amount of pharmaceuticals in the environment due to new waste uses and treatments may have several consequences. First of all, legislation dealing with these waste streams may need to be adapted, especially in case of an increased load. Secondly, the currently used approaches for exposure modelling of pharmaceuticals in the environmental risk assessment in the marketing authorisation process may need to be revised.

The aim of this report is to further identify those routes, where the chance of pharmaceuticals entering the environment is changing when compared to the conventional waste treatment or use. The possible implications for legislation and/or marketing authorisation of pharmaceuticals will only be discussed briefly.

Traces of pharmaceuticals may not only enter the environment, but also end up in the recovered raw material. Waste materials must be reclassified (*i.e.* no longer legally labelled as 'waste') before they can be converted into raw materials or products. This means that the relevant waste stream must meet specific safety and technical requirements to ensure that its reuse does not result in unacceptable risks to human health or the environment. These requirements are referred to as 'end-of-waste' criteria (for background details: Spijker and Van der Grinten, 2014). In this report we will not go into detail on policy steps to further promote the reuse of waste streams towards a circular economy. We will highlight, however, some aspects that are important when weighing sustainability and safety criteria in these new waste-to-product chains (paragraph 4.2).

The most important technological developments will be addressed, but since the developments and changes in routes/techniques go very fast, this overview cannot be exhaustive. Developments which are still in the exploratory lab scale phase are not considered. Waste due to production and transport of pharmaceuticals is not taken into account in this report.

Wherever the term 'pharmaceuticals' is used in the report, both the parent compound and possible metabolites are meant. In this report

pharmaceuticals are assumed to be organic substances. This is however not always the case: some pharmaceuticals contain metals like zinc, which are by nature not degradable. Throughout treatment of waste streams these metals might concentrate, for example during digestion or composting. This aspect is not taken into account in detail.

The presence and promotion of pathogens, antibiotics and microbial resistance genes is an aspect that is worth attention, but beyond the main scope of this report.

Finally it should be emphasised again that the waste streams that are considered in the report (*i.e.* urine, faeces, wastewater, sewage sludge and manure) will not only contain pharmaceuticals, but also many other micropollutants, for example cleaning agents or personal care products. Many remarks that are made on pharmaceuticals may also apply to these micropollutants (see also paragraph 1.1). They are however not considered in further detail in this report.

Chapter 2 and 3 will present sanitation and re-use possibilities for waste streams containing, respectively, human and veterinary pharmaceuticals. The potential impact on the environmental load of pharmaceuticals from these routes will be discussed. Chapter 4 focusses on the general conclusions and recommendations.

2 Sanitation and reuse of waste containing human pharmaceuticals

2.1 Leftover medication entering the environment

For the Netherlands in 2013 (Reitsma et al., 2013), 54% of respondents in a survey took the unused pharmaceuticals back to the pharmacist who collected them. About 6% of the respondents took the medication to the pharmacist, but the pharmacist refused to take the medicines back. Nearly 11% of the respondents put the unused medicines in the waste bin, while 2% flushed the unused medicines through the toilet (list is not exhaustive, please refer to Reitsma et al for the complete list).

It is unknown whether the amount of unused pharmaceuticals is increasing. When leftover pharmaceuticals are put in the waste bin, the environmental load is considered low since in the Netherlands all waste (either from the pharmacist or the waste bin) is incinerated. However, unused medicines that are flushed through the toilet will end up in the sewage treatment plants (STPs), where they are partly removed (see paragraph 2.2). Currently, the Dutch ministry of Infrastructure and Environment is making detailed agreements with pharmacists about the collection of leftover medication. These (non-technological) actions may eventually result in a decrease of the environmental load of drugs. This topic is not further discussed here.

2.2 Wastewater

2.2.1 *Conventional wastewater treatment*

The objective of conventional STPs was to produce environmentally safe wastewater by removing nutrients and solids. Since then, most wastewater treatments have improved and currently also include activated sludge treatment. This treatment partly removes micropollutants.

Conventional wastewater treatment often consists of a number of steps, including primary treatment where solids can settle and secondary treatment using activated sludge to remove organic compounds. Within this secondary treatment, a treatment can be introduced with a number of adjustments like anaerobic tanks, to also remove nutrients.

The resulting wastewater stream is then emitted onto surface waters, while the sludge can be digested, incinerated, used as filling for cement or as composted (Compendium voor de Leefomgeving, 2014).

2.2.2 *Upgrading of sewage treatment plants*

Several concepts and post-treatment techniques to improve removal of micropollutants (including pharmaceuticals) from wastewater have been developed in the past years, *i.e.* membrane bioreactor (MBR), ultrafiltration, 1-step filter, packed or granulated activated carbon, advanced oxidation processes (ozone, UV-H₂O₂ or TiO₂) with or without subsequent activated carbon filtration, and more.

The status of these concepts and techniques differs, from experimental via pilot scale to full scale implementation. In the Netherlands some full scale membrane bioreactor plants have been operated for several years but are now closed down (Schyns et al., 2012). A full scale 1-step filter is operational (STOWA, 2013a). Pilot studies have been performed with among others activated carbon (STOWA, 2009; STOWA, 2010) and advanced oxidation techniques (STOWA, 2009). In Switzerland¹ and Germany² advanced treatment techniques have been investigated extensively and several have been implemented at STPs on full scale.

In general, all these techniques improve the removal of pharmaceuticals to a certain extent, although they are not equally efficient and the amount of transformation products (metabolites) with unknown properties may increase. Advanced oxidation processes seem to be most efficient, although in some cases toxic metabolites may be formed. By changing the process conditions and/or installing a subsequent activated carbon filtration these degradation products can be eliminated.

In the following paragraph we will discuss the 'evolution' of these sewage treatment plants towards production plants for energy and other products.

2.3 Sewage treatment plants as a factory for energy and other products

2.3.1 Background

In 2012, the Association of Regional Water Authorities (in Dutch: Unie van Waterschappen) and the Association of Netherlands Municipalities (Vereniging Nederlandse Gemeenten) presented their vision on future developments in the wastewater chain (Römgens et al., 2012) in which they describe their contributions to the development of a more sustainable society. Their basic assumption is that treatment of wastewater is no longer based on destruction, but on regaining of resources and energy. The ambition of the water boards is that they generate at least 40% of their energy needs by themselves in 2020. Furthermore the possibilities to (re)generate organic material from wastewater are investigated. This development fits very well within the ambition of the Dutch government to promote a circular economy (see Chapter 1).

A wide range of projects has been started up and implemented (for an overview: see www.energiefabriek.nl and www.grondstoffenfabriek.nl, both in Dutch).

The major products that can be regained from wastewater are biogas, phosphate, bioplastic, cellulose, alginate and biomass. Other potential products have been considered as well, such as nitrogen from the rejection water, the concentration of COD for the production of fuel and

¹ MicroPoll project, <http://www.bafu.admin.ch/>; topics; water protection, micropollutants.

² Milieuministerie Nordrhein-Westfalen (MKULNV) project 'Elimination von Arzneimitteln und organischen Spurenstoffen', www.micropollutants.net (12 projects, different techniques, also full scale)

the recovery of CO₂ from biogas. This CO₂ can be used in greenhouses to increase the production or in the food and drinking water industry. However, it was concluded that at this stage it is very expensive to recover these additional potential resources, or the possibilities are (still) hampered by legislation (STOWA 2013b).

An important point of attention is that for any new product regained from wastewater, there needs to be a moment when the product is no longer labelled as 'waste'. For this the product has to comply with certain technical criteria (considering the quality of the product), to be safe for the environment and human health, the material has to be commonly used for specific purposes and there has to be a market for the product. These criteria are called the End of Waste criteria (see also paragraph 1.4). For all resources mentioned above except biogas, these End of Waste criteria still need to be established. Guidance how to deal with End of Waste criteria is given in Spijker and Van der Grinten (2014). Some legal aspects of the transition of STPs to resource factories have also been addressed in STOWA (2012a).

An overview of the energy and other products that can be obtained from sewage treatment plants is given in Table 1. A more detailed description is found below, including an indication on the potential changes in the environmental load of pharmaceuticals.

2.3.2 *Biogas*

About 50% of the primary and secondary sludge of the STPs in the Netherlands is digested (STOWA, 2011b), at about 80 locations (Van der Wal, 2014)³. Primary and secondary sludge (settled pre and post activated sludge treatment, respectively) are usually treated together. In the sludge digestion tank the organic substances are converted into methane (biogas), water and carbon dioxide, while a smaller volume of digested sludge remains. The supernatant water is lead back to the STP as rejection water. The methane can be used as biogas and hence serve as an energy source. Digestion reduces not only the volume of the sludge, but also makes it more stable and more easily to dewater, which all reduces the treatment costs for the remaining sludge.

Three different processes / possibilities can be distinguished:

1. Mesophylic digestion
This is the conventional sludge digestion method. It takes place in a digestion tank at a temperature of 30 to 38 °C and a sludge retention time of about 20 days (STOWA, 2011b).
2. Thermophylic digestion
Digestion at a temperature of around 55°C. Thermophylic digestion results in a degradation of organic matter and a higher biogas production than mesophylic digestion (STOWA, 2012c; STOWA, 2014c).
3. Thermal hydrolysis
Prior to the digestion the sludge is pretreated under high pressure (6-8 bar) and high temperature (up to 170°C). This way

³ There were 343 STPs in the Netherlands in 2012 (CBS, 2014). At some locations the sludge of multiple STPs is digested.

the organic matter is better degraded in the digestion, leading to a higher biogas production (STOWA, 2011e; STOWA, 2012b). The process is called thermal hydrolysis. Pretreatment by thermal hydrolysis can be applied to mesophylic as well as thermophylic digestion

The fate of pharmaceuticals during these anaerobic digestion processes has hardly been studied. Carballa et al. (2007) studied the removal of some antibiotics and other pharmaceuticals during digestion under mesophylic and thermophylic conditions and with different Sludge Retention Times (SRTs). In general, no influence of SRT and temperature on pharmaceutical removal was observed.

In general most pharmaceuticals are not readily degraded under anaerobic conditions (Kujawa-Roeleveld, 2008). When organic matter is degraded more easily than pharmaceuticals this might lead to a concentration of pharmaceuticals in the remaining sludge. Since in the Netherlands this remaining (digested) sludge generally is incinerated, this has no consequences for the load of pharmaceuticals to the environment.

Table 1 Energy and other products which can be obtained from sewage treatment plants. The load refers to the potential impact on the emission of pharmaceuticals to the environment.

Product	Where in STP	How?	Status	Use of product	Expected extent	Pharmaceuticals?	Load?	References
Biogas	Primary and secondary sludge	Mesophylic digestion	Operational at about 80 STPs	Energy	Increase (more 'energiefabrieken'; Klimaatakkoord UvW http://www.uvw.nl/publiecatie/klimaatakkoord-unie-van-waterschappen-rijk/)	Not known	Not known	STOWA (2011b) STOWA (2006)
	Primary and secondary sludge	Thermophylic digestion	Operational at STP Echten, research at STP Bath and Leeuwarden	Energy	Increase (more 'energiefabrieken'; Klimaatakkoord UvW http://www.uvw.nl/publiecatie/klimaatakkoord-unie-van-waterschappen-rijk/)	Not known	Not known	STOWA (2012c); STOWA (2014c) STOWA (2006)
	Primary and secondary sludge	Thermal hydrolysis as pretreatment for mesophylic or thermophylic digestion	Operational at STP Venlo and Apeldoorn (full scale), pilots in 2015 at STP Echten and Tilburg	Energy	Not known	Not known	Not known	STOWA (2011e); Hol et al. (2014)
Phosphate	From sludge or rejection water (from sludge dewatering and/or sludge digestion)	Precipitation with magnesium. Different processes (Pearl, Airprex, Anphos)	Operational at 5 STPs, planned in near future at 10 STPs	Fertilizer (replacing conventional fertilizer)	Not possible for STPs with chemical P-removal. Potential for broad implementation is high. The legislation is an obstacle for use, but initiatives for approval as fertilizer are ongoing.	Different processes lead to different purity of struvite. The uptake of pharmaceuticals is currently verified (STOWA, in prep).	Not known yet, results foreseen in 2015	STOWA (2011a); Ehlert et al. (2013a); van Veldhoven (2015)

Table 1 CONTINUED Energy and other resources which can be obtained from sewage treatment plants. The load refers to the potential impact on the emission of pharmaceuticals to the environment.

Product	Where in STP	How?	Status	Use of product	Expected extent	Pharmaceuticals?	Load?	References
Phosphate	From ashes (after incineration of sludge)	Recover from ashes, different processes (Ashdec, SNB-Thermphos, Aliphos) and different P-products	Full scale project starting at EcoPhos in Vlaardingen	Fertilizer (replacing conventional fertilizer)	Potential is high. The legislation is an obstacle for use, but initiatives for approval as fertilizer are ongoing.	Not known. Different processes are expected to lead to different purity of P-product	Not known	STOWA (2011a); Ehlert et al. (2013a); HVC et al (2015)
Bio-plastic (PHA)	Primary sludge and secondary sludge	Micro-organisms produce PHA-polymers from volatile fatty acids (which are produced by acidifying sludge)	Explorative	Degradable plastics (for agricultural foils, packing materials, consumer products)	Demand for PHA is increasing; costs need to be reduced to be commercially competitive.	Not known	Not known	STOWA (2014d)
Cellulose	Influent	From toilet paper that is sieved from the influent	Principle proven, full scale sieving of influent in about four Dutch STPs	Raw material for the production of asphalt, isolation materials and fibre strengthened synthetic materials	High potential, competitive with recycled paper.	Not known; some sorption to organic matter may be expected	Not known	STOWA (2013d)

Table 1 CONTINUED Energy and other resources which can be obtained from sewage treatment plants. The load refers to the potential impact on the emission of pharmaceuticals to the environment.

Product	Where in STP	How?	Status	Use of product	Expected extent	Pharmaceuticals?	Load?	References
Cellulose	Sewage sludge	From cellulose fibres that is filtered from the sewage sludge	Pilot/Lab scale	Raw material for the production of asphalt, isolation materials and fibres strengthened synthetic materials	Depending on lab scale, results expected in 2015	Not known; some sorption to organic matter may be expected	Not known	STOWA (2014a)
Alginate	Granular biomass of NEREDA installations	Extraction of the granular biomass	Lab scale, pilot in preparation	Various: see http://www.stowa.nl/projecten/Alginat_teru_gwinnen_uit_korrelsib	Results of feasibility study expected in 2015	Not known	Not known	STOWA project 432640
Biomass	Effluent	Growth of algae, duckweed, water plants, willows, mussels etc. on effluent	Explorative	Energy, algae based products, food for animals	Increasing, extent not clear	Not investigated, some uptake in biomass is to be expected	Depends on use of biomass: when used for biogas production no load, when used as food or in algae based products increased load	Otte & van Hoorn (2014); Otte et al (2014); STOWA (2011d)

On the other hand, pharmaceuticals that were removed in the STP by sorption to activated sludge might desorb again during digestion. Part of these pharmaceuticals will be led back to the STP together with the rejection water, a part will be degraded and a part will remain in the digested sludge, which is incinerated. Whether the load that is led back with the rejection water is significant or not is not known, a mass balance has not been made. Two pilot projects in which the release of micropollutants from the sludge will be measured are ongoing at, respectively, STP Echten and STP Tilburg.

2.3.3 Phosphate

In STPs phosphate is removed from the raw wastewater either chemically (by precipitation onto the sludge), biologically (by bacteria) or by a combination of both. Every year 11.000 to 12.000 kg phosphorus ends up in the sewage sludge (STOWA, 2011a) and is then incinerated. Since phosphate is a valuable resource, processes have been initiated to recover the phosphate, by precipitation with magnesium, forming struvite ($\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$). In this way also part of the nitrogen is removed. Struvite can be used as a fertilizer.

There are two major stages at which struvite can be recovered: 1) at the STP, from sewage sludge or rejection water; 2) from the ashes remaining after incineration of sewage sludge. Several processes have been compared in STOWA (2011a) and Eekert et al. (2013), where it is shown that these different processes also lead to different quality (*i.e.* purity) of struvite.

From April 2015, struvite has been approved as fertilizer⁴. An important issue is whether or not substances (such as pharmaceuticals) are incorporated in the product. When struvite is used as a replacement of conventional fertilizers the load of pharmaceuticals into the environment might increase. The uptake of pharmaceuticals (and other micropollutants) is currently verified (STOWA, in prep).

2.3.4 Bioplastics

The expression 'bioplastic' is used for biological degradable plastics and/or plastics produced from a renewable resource. Plastics produced from a renewable resource hence are defined as bioplastics, but are not necessarily biodegradable.

Some bioplastic types are produced by micro-organisms using a carbon source as substrate. PHA (polyhydroxyalkanoate) is such a bioplastic. It is produced by using volatile fatty acids as a carbon source. These volatile fatty acids can be regained from sewage sludge by acidifying the sludge. The technical and economic potential to produce PHA bioplastic from sewage sludge has been explored in STOWA (2014d). It was concluded that the production of bioplastic from sewage sludge is technical feasible, but the costs are still high and several aspects still need to be investigated and improved. In June 2015 several parties signed an agreement on the production of degradable bioplastics from sewage sludge at Bath, the Netherlands (see: http://www.stowa.nl/nieuwsagenda/nieuws/van_afvalwater_naar_biologisch_afbreekbaar_plastic).

⁴ http://wetten.overheid.nl/BWBR0019031/geldigheidsdatum_23-04-2015#BijlageII

It has, to our knowledge, not been investigated whether pharmaceuticals (or other micropollutants) end up in the bioplastic.

2.3.5 *Cellulose*

Influent from the STP contains a lot of toilet paper, which is rich in cellulose. About 35 to 40 percent of the suspended matter in wastewater consists of cellulose fibres (Kampschreur, 2014). Cellulose can be used as raw material in a number of products, for example in asphalt as a binding material, in isolation materials, in synthetic materials as a fibre strengthener, in paper and cardboard and to make bioplastics. The cellulose can be recovered from the influent by fine sieving (STOWA, 2013d). Furthermore pilot/lab scale testing facilities have been initiated to recover the cellulose from sewage sludge (STOWA, 2014a).

The reuse of cellulose from STPs is technically very well feasible, and the costs are competitive compared to other sources for cellulose, such as recycled paper (STOWA, 2013d). Theoretically, 150.000 tonnes of cellulose per year can be regained from STPs in the Netherlands (STOWA, 2014a). It has not been investigated whether pharmaceuticals (or other micropollutants) end up in the cellulose. Prior to use the cellulose needs to be treated to clean it from other materials that are removed by fine sieving and to make it hygienically safe (pathogens), for example by autoclaving. It has not been investigated whether pharmaceuticals adsorb to the cellulose, and what is the influence of the clean-up and further treatment of sieved material. Some adsorption may be expected.

2.3.6 *Alginate*

Recovery of alginate is interesting for STPs which use the Nereda technology. The Nereda-technology is a compact wastewater treatment technique with so-called aerobic granular biomass: purifying bacteria that create compact granules with very good settling properties.

Research has revealed that the substance that creates these bacterial granules is an alginate polymer. The granules contain up to 20% alginate, which is more than twice as much as conventional sludge (STOWA, 2013c). Alginate is a sugar like substance that attracts a lot of water, thickens or jellifies fluids and can form the basis for coatings. It has a lot of possible uses: see http://www.stowa.nl/projecten/Alginaat_terugwinnen_uit_korrelsib. After extraction of the alginate the remaining bacterial granules are ready to be digested, hence producing biogas.

The possible trading markets are being investigated in the project 'Recovery of alginate from sludge granules' (STOWA-project 432640). It has not been investigated whether pharmaceuticals are taken up in the product.

2.3.7 *Biomass*

Several projects have been initiated to grow algae (STOWA, 2011d), duckweed (Otte & van Hoorn, 2014), willows (Otte et al., 2014) as well as mussels (ref: mosselexperiment Rijn en IJssel) on STP effluent. Goal of this post-treatment is to further remove nutrients.

The biomass recovered from this biological post-treatment could serve a resource for valuable products such as oils, proteins or fatty acids from

algae, animal food, or glue and coatings from the proteins from duckweed. The biomass may also be used in (co)digestion to produce biogas.

So far, post-treatment of effluent by growing biomass, has not made it beyond pilot scale, but this may change in the future. It can be expected that during the treatment some degradation as well as uptake of the pharmaceuticals in the biomass may take place. This will lead to a better effluent quality, thus lowering the risk to the environment. However, the pharmaceuticals in the biomass may enter the environment through other routes, *i.e.* in products and/or by the rejection water from the digester (see paragraph 2.3.2).

2.4 New sanitation concepts

2.4.1 Background

In domestic wastewater four basic water streams can be distinguished (STOWA, 2008):

- Feces (or with water diluted feces): brown water
- Urine (or with water diluted urine): yellow water
- Water originating from bath, shower, washing machine and kitchen: grey water
- Rain water: blue water

Water originating from toilet flushing is sometimes also referred to as black water. Black water contains urine and feces, diluted with water used to flush the toilet.

These water streams are of different composition. Urine and feces contain most of the nutrients (N and P), as well as most, if not all pharmaceuticals, which are concentrated in only about 1% of the volume of wastewater that a person produces daily (see Figure 3).

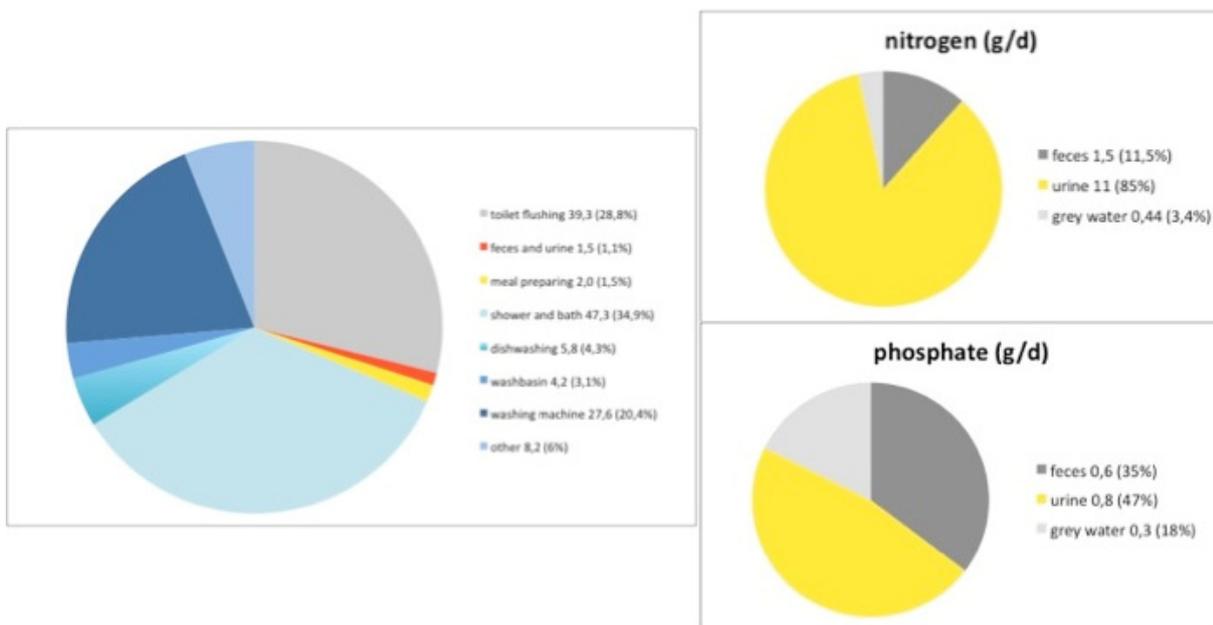


Figure 3 Volume of domestic wastewater in litres/day, and in %, and the origin of nitrogen and phosphorus in domestic wastewater in gram/day and in % (STOWA, 2008).

In the conventional situation, all water streams are combined, and treated together. This way wastewater is strongly diluted, making the wastewater treatment less efficient. New concepts have been initiated to treat wastewater streams in a different way, while at the same time nutrients and energy are recovered. These are called 'new sanitation concepts'. They might be cheaper, more sustainable and more effective. A common feature is that wastewater streams are treated in other combinations, in some cases also together with other waste streams such as food waste. There are many different concepts, adapted to local circumstances.

In the period from 2000 until now many pilots have been performed. In September 2014 a demonstration site opened at the Antonius hospital in Sneek, where three wastewater streams are available for tests and pilots: hospital wastewater and communal urine and black water. Currently several new sanitation concepts go towards implementation. These are described in the following paragraphs and summarized in Table 2. Since grey water contains little or no pharmaceuticals, separate treatment of grey water is not further elaborated.

2.4.2 *Urine separation (yellow water)*

Urine contains about 85% of the nitrogen and 47% of the phosphorus in urban wastewater (STOWA, 2008). Besides this, it also contains most of the excreted pharmaceuticals and hormones. These substances are concentrated in only about 1% of the total amount of wastewater daily produced by one person, *i.e.* in little over 1 litre per day (STOWA, 2008; see also Figure 3). This makes it in theory attractive to collect urine separately, regain nutrients and remove pharmaceuticals and hormones. At the same time it also decreases the load of nutrients, pharmaceuticals and hormones discharged to the STP.

GMB BioEnergy developed the first full scale urine treatment factory, called SaNiPhos⁵. The urine is originating from situations where it is already collected separately such as pop music festivals and 'Mothers for mothers' (a urine collection system for pregnant women). In SaNiPhos phosphorus (and partly also nitrogen) is precipitated with magnesium, forming struvite. Nitrogen is further removed by forming ammonium sulphate. Both struvite and ammonium sulphate can be used to replace conventional fertilizer (see 2.3.1. for current legal restrictions). The treated urine (with pharmaceuticals) is discharged to the sewage system. Since the pharmaceuticals are not removed and still end up in the sewer, there is no change in their environmental load.

There is however a pilot ongoing to further treat the urine with activated carbon to remove pharmaceuticals and hormones (GMB, 2014). If this further treatment will become practice this will lower the load of pharmaceuticals to the environment.

Other options to treat separated urine have also been investigated on pilot scale. In the village of Anderen urine was directly used on the land as fertilizer. This pilot did not get any follow-up due to multiple reasons. In the city of Sneek urine was treated with ozone and activated carbon to remove pharmaceuticals and hormones. Also this pilot did not get any follow-up, despite the fact that the treatment with ozone and activated

⁵ <http://www.gmb.eu/SaNiPhos.aspx?NL-1-289-0>

carbon was very effective. The most important reasons were problems with the collection of urine, *i.e.* clogging of the pipelines of the urine separation toilets, the treatment costs and safety matters with regard to the ozone generator (Wortel, 2014).

In further initiatives to explore the possibilities to collect urine separately, the logistic matters and integration of the system within buildings have turned out to be an important obstacle. For this reason a broad implementation of urine separation is not expected.

2.4.3 *Separate treatment of black water*

In 2005 a total of 32 houses in the area Lemmerweg Oost in the city of Sneek have been equipped with vacuum toilets to collect the black water separately. This black water is digested in an Upflow Anaerobic Sludge Bed reactor (UASB). This is a small decentralised digester that treats the black water from the housing area only. This way biogas is produced, which is used to generate energy, thus lowering the energy consumption to 50% compared to conventional systems. After digestion phosphorus and a part of the nitrogen are recovered as struvite, followed by further nitrogen removal. The effluent is discharged to the conventional sewer system (STOWA, 2014e). Besides this, grey water and rainwater are also collected separately, but since these water streams do not contain pharmaceuticals they are not further elaborated here. The operation and performance of the system have been evaluated in STOWA (2014e). The effluent of the treated black water is not clean enough (in terms of nutrients) to meet criteria for direct discharge to the surface water. Research with regard to the effectiveness of the system to remove pharmaceuticals (and synthetic and natural hormones, which can act as endocrine disruptors) is ongoing.

The black water collected at Lemmerweg Oost was also used in a study of de Graaff et al. (2011). The black water was first anaerobically treated in a UASB (Upflow Anaerobic Sludge Blanket) reactor. The effluent of the UASB reactor was subsequently treated in a two reactor nitrification-anammox process to remove the nitrogen. The fate of 11 pharmaceuticals (and some hormones) were followed during this process. Only paracetamol was removed anaerobically. Some of the other pharmaceuticals were partly removed in the nitrification-anammox process. It was concluded that the performed biological treatment alone is not enough to eliminate the selected compounds from black water.

To our knowledge, the extent to which black water is treated separately, is very limited in the Netherlands, apart from two cases in which black water is treated together with organic waste (see paragraph 2.4.4). Apart from that the Dutch Institute for Ecology (NIOO-KNAW) in Wageningen has plans to grow algae on the digestate, *i.e.* the water fraction of the digested black water. So far only tests at a lab scale have been performed and for this reason this pilot is not further elaborated here (Fernandes, 2013).

Table 2 New sanitation concepts. The load refers to the impact on the emission of pharmaceuticals to the environment.

Waste (water) stream(s)	Name / location	Products	Status	Use of product	Expected extent	Pharmaceuticals?	Load?	References
Urine	SaNiPhos	Struvite, ammonium-sulphate	Operational	Fertilizer	One site in NL. Currently no plans for other locations.	Not removed	Equal to conventional (lower if effluent is further treated with activated carbon)	GMB (2014)
Black water	Lemmerweg Oost in Sneek	Biogas, struvite	Operational	Energy, fertilizer	One site in NL. Currently no plans for other locations.	Not known, research ongoing, some removal can be expected	Decreased when effluent is discharged to sewer, increased when discharged directly to surface water	STOWA (2014e)
Black water, organic waste, grey water	Noorderhoek area in Sneek	Biogas, struvite, heat	Operational	Energy, fertilizer	One site in NL. Currently no plans for other locations.	Not known, some removal can be expected	Decreased when effluent is discharged to sewer, increased when discharged directly to surface water	STOWA (2014b)
Black water, organic waste, STP sludge	Autogeneratieve High Pressure Digestion	Biogas, CO ₂ , phosphor	Explorative	Energy, fertilizer,	Pilot at one site in NL. Negotiations for demonstration project ongoing	Not known, some removal can be expected	Decreased when effluent is discharged to sewer,	Zagt (2015)
Black water, hospital waste, organic waste	Pharmafilter	Biogas	Operational and planned	Energy	7 locations operational and/or planned, number increasing	Completely removed	Reduced to zero	STOWA (2013e)

When the effluent of the anaerobic black water treatment is discharged to the conventional sewer, the load of pharmaceuticals will - to some unknown extent - be lower than in the conventional system. When algae are grown on the effluent, part of the pharmaceuticals may be taken up by the algae. Further pharmaceutical exposure routes depend on the subsequent application of the algae and the remaining effluent.

2.4.4 *Combined digestion of black water and organic waste*

Black water can be digested together with organic waste, *i.e.* vegetable and fruit remains. Two concepts on a residential area scale are described below. In these concepts, besides combined digestion of black water and organic waste, also energy saving is an important aim.

Furthermore it is also possible to place an organic waste grinder in the kitchen, hence transporting the organic waste (*i.e.* vegetables, fruit and other organic kitchen remains) with the wastewater to a STP for central treatment (NOS, 2014). This concept is not further elaborated since it still is in a preliminary phase.

In a discussion of the 'Platform new sanitation'⁶ on April 16th 2014, it was concluded that the combined digestion of black water and organic waste certainly has potential, but logistic challenges have to be solved. Pilots must learn whether it is feasible or not in practice (Swart, 2014).

Integral energy concept: Noorderhoek area in Sneek

In the newly built Noorderhoek residential area in Sneek a completely new concept for collection, transport and treatment of wastewater and organic waste (vegetable and fruit remains) has been implemented and tested (STOWA, 2014b). In total 232 houses are foreseen to be built in this area, but due to stagnation on the housing market only part of these have been realized (and tested) so far.

Black water and organic waste (vegetable and fruit remains) are collected for the whole area and led together to a digester, *i.e.* an UASB reactor, where the organic material is converted to biogas. Furthermore phosphate is regained by forming struvite. Grey water is also treated separately. Furthermore the heat of the grey water as well as the biogas are used by a central installation that warms water for a low temperature heating system for the whole area. The goal is that the treated effluents (from the digester and the grey water treatment) are clean enough to discharge directly to the surface water, but due to the experimental character the effluent is currently discharged to the sewer system (STOWA, 2014b).

The removal of pharmaceuticals in the black water treatment has been investigated only to a limited extent: eight pharmaceuticals were measured a few times. Of these, five were removed for >95%, one for 87% and two with lower efficiencies (68 and 16%) (STOWA, 2014b). These high removal efficiencies seem to be in conflict with the results of Kujawa-Roeleveld (2008) and De Graaff et al. (2011) who found much lower removal of pharmaceuticals under anaerobic conditions. Maybe the presence of organic waste plays a role as co-digester that improves the degradation process.

⁶ A network of professionals dealing with new sanitation concepts.

When the effluent of the anaerobic black water treatment is discharged to the conventional sewer, the load of pharmaceuticals will - to some unknown extent - be lower than when untreated wastewater is discharged from households in the conventional sewer system. When the effluent is directly discharged to the surface water, this might increase the emission of pharmaceuticals to the surface water. In this case further verification of the performance of the digester will be necessary.

Autogenerative High Pressure Digestion

Autogenerative High Pressure Digestion (AHPD) is a process for digestion of organic material that is derived from the food industry. It can be used to digest different types of organic matter. The digestion takes place in a closed anaerobic reactor at high pressure (20 bar) and a temperature of 35 - 55°C. Both pressure and temperature can be regulated. The high pressure is generated by the production of biogas: methane will not dissolve in water so the more methane is produced the higher the pressure will be. At this high pressure the CO₂ will dissolve, leaving a biogas with a high methane content that can directly be used without further purification. The water phase is drained batch wise and ultrafiltered. The concentrate is lead back to the reactor. The permeate (*i.e.* the water phase after ultrafiltration) contains dissolved CO₂, H₂S and NH₃ gasses. These gasses will release from the permeate causing the pH to rise up to 13. Due to this high pH phosphate in the water will precipitate with the metals in the water, hence removing about 50% of the load. In summary the system produces high quality biogas (about 90% CH₄), CO₂ which for example can be used in greenhouses to increase the production, a phosphate precipitate, water with relatively low metal and phosphate content and digested sludge which will be incinerated (Zagt, 2015; www.bareau.nl).

It is claimed that the AHPD technology can be used in all processes in which biomass has to be treated, such as wastewater, sewage sludge and waste streams from agriculture and food industry. When used to treat wastewater, the minimum scale to be cost effective is at the scale of a residential area.

The system has been operated for three years on a pilot scale, processing different mixtures of black water, kitchen waste from a restaurant, sludge from the STP Drachten and/or glycerol from a biodiesel factory. The next step will be a demonstration project. Negotiations for such a project at a small STP are in an advanced stage (Zagt, 2015).

The influence of the process on pharmaceuticals (or other micropollutants) has not been investigated. Some degradation may be expected in the reactor. Furthermore the high pH might influence the stability and behavior of the pharmaceuticals. The influence of high pressure on the pharmaceuticals is unknown.

When the effluent of the reactor is discharged to the conventional sewer, the load of pharmaceuticals will - to some unknown extent - be lower than when untreated wastewater is discharged in the conventional sewer system.

2.4.5 *Treatment of hospital wastewater*

Hospital wastewater contains 5-10% of the total pharmaceutical load, concentrated in about 0.4% of all wastewater (Derksen & Ter Laak, 2013). The concentrations of pharmaceuticals are relatively high, but besides that the risk of hospital wastewater is higher than of domestic wastewater due to the more potent pharmaceuticals that are used in hospitals (Nafo et al., 2012; Anonymous, 2011) and the possible presence of antimicrobial resistant bacteria. This makes hospital wastewater an interesting target for separate treatment. Different advanced wastewater treatment techniques to treat hospital wastewater have been investigated and have demonstrated to be effective, on lab scale, pilot scale as well as full scale in several hospitals along Europe (among others in the PILLS project: Nafo et al., 2012). By nature these techniques all lead to removal of pharmaceuticals, and hence to a reduction of the risk. A major drawback however to implement these techniques are the high implementation and maintenance costs. It is therefore not expected that these techniques will be implemented to a high degree. An exception is the Pharmafilter concept, which is further elaborated below.

Pharmafilter

Pharmafilter is an integral concept to optimize logistics in care processes and waste streams, and treat waste and wastewater within hospitals. The unique feature of the Pharmafilter concept is that the costs of the on-site wastewater treatment are covered by savings in logistics for the nurses, as well as handling and removal costs for waste streams. Along with this, also safety and hygiene are increased, resulting in decreasing infection rates. For the hospital this has hygienic, logistic and financial benefits.

In hospitals, feces and urine are often collected in bedpans and urinals. The disposal of the feces and urine, as well as the cleaning of the bedpans and urinals may lead to unhygienic situations. In the Pharmafilter concept feces and urine are collected in single use products made from bioplastic. After use these are shredded in the Tonto (a specially designed shredder) and discharged together with the wastewater to the internal sewer system of the hospital. Also other organic waste such as food waste is shredded in the Tonto and removed this way.

The mixed waste stream is led to a central installation within the hospital complex. The mixed waste stream is separated in solid and liquid waste. The solid waste is anaerobically digested, thereby producing biogas. This biogas is used onsite to provide energy to the water purification installation. The liquid waste (*i.e.* wastewater) is treated in a membrane bioreactor (MBR) with ultra-filtration membranes. Effluent (permeate) from the MBR is further treated with ozone and activated carbon. The effluent is discharged to the sewer. The interaction between the MBR and the digester is innovative. The sludge discharged from the MBR is fed back into the digester and any excess water from the digestate (*i.e.* water phase) of the digester is treated in the MBR. The digested sludge is disposed of in traditional ways (*i.e.* it will be incinerated).

The performance of the concept was first tested on a pilot scale at the Reinier de Graaf Gasthuis in Delft. Since October 2010 a full scale installation is in use. An extensive monitoring of pharmaceuticals and other micropollutants has been carried out. Although high concentrations of pharmaceuticals were present in the influent of the MBR, no pharmaceuticals could be detected in the effluent (*i.e.* after activated carbon treatment) (STOWA, 2013e).

It can be concluded that the Pharmafilter concept decreases the emission of pharmaceuticals to the sewage at the hospital site to nihil. This will decrease the risk of emission of pharmaceuticals to the surface water. So far seven Pharmafilter installations are operational and/or planned in the Netherlands, and the number is extending (Wortel, 2014).

3 Sanitation and reuse of waste streams containing veterinary pharmaceuticals

3.1 Manure treatment

3.1.1 Background

The Dutch policy is aimed at reducing the emission of nutrients to the air, water and soil. This could partly be achieved by replacing chemical fertilizers with (products from treated) manure (MinEZ, 2011). On January 1st 2014, the Dutch Law on Fertilizers ('Meststoffenwet') has been amended. Since then farms that cannot apply all of their manure on their own fields (based on phosphate), are obliged to partly treat this surplus-manure. For 2015 the percentage of manure that has to be treated is established at 50% for region South, 30% for region East and 10% for the other Dutch regions (MinEZ, 2014). The aim of this policy is that the manure, after manure treatment, is used outside the Dutch agriculture, thus finding an answer to the manure surplus.

Manure treatment also aims to separate the manure in different phases and/or (mineral) concentrates: nitrogen and potassium are mainly found in the liquid phase, while phosphate mainly remains in the solid phase. This separation of manure in different phases or concentrates offers the advantage of more precise fertilization options and less transport movements.

In Table 3, techniques for treatment of manure are listed. The table is limited to techniques that have proven themselves in practice. Separation of manure in a liquid and a solid phase is a pre-treatment for several other treatment techniques. It is only discussed in paragraph 3.1.2, and referred to in other paragraphs. Mixing of manure is not discussed. Burning of manure is not discussed either: it will remove all organics including pharmaceuticals, thus eliminate the emission (even when the ashes are used).

Several estimates are available on the number of installations in which manure is treated. CBS (2015) estimates about 20 installations, Korving et al (2013) about 140, while another inventory of the manure treatment capacities gives a number of 67 operational installations (Verkerk et al, 2014). Details of a very recent national inventory can be found via:

<http://www.mestverwerkingsloket.nl/Static/Documents/UserUpload/Rapportage%20Landelijke%20Inventarisatie%20Mestverwerking%202014.pdf>.

Another aspect that needs to be mentioned is that for the use of (mineral) concentrates as replacement of chemical fertilizer, the legislation has to be adapted. With the permission of the European Commission ten pilots have been operated since 2009 to test the feasibility and effects of mineral concentrates as replacement for chemical fertilizer (the most recent information can be found via: <http://www.wageningenur.nl/nl/Publicatie->

[details.htm?publicationId=publication-way-343930333231](#)). The results are used to get a permanent permission from the EC (Korving et al, 2013). The recognition of manure treatment installations for production of a mineral concentrate (paragraph 3.1.2) is still pending (Van der Hulst, 2015).

The current legislation prescribes the maximum amount of nutrients (e.g., nitrogen) that may be applied on the land (MinEZ, 2014). When, during the various manure treatment processes, the amount of pharmaceuticals does not change relative to the amount of nutrients, no increased load of pharmaceuticals is to be expected.

3.1.2 *Manure treatment installations for production of mineral concentrate*

In manure treatment installations the manure (mostly pig manure) is separated in a solid phase, a mineral concentrate and a permeate (effluent). Three treatment steps can be distinguished:

1. Separation in a liquid and a solid fraction
Several methods are applied: filtering, sieving, settling or centrifugation. Also a filter belt press is used, after flocculation with polyelectrolyte.
2. Pre-treatment
The liquid phase is further dewatered by ultrafiltration, nanofiltration or dissolved air flotation. This dewatering step reduces the volume that needs to be treated by reverse osmosis.
3. Reverse osmosis
The pre-treated liquid phase flows under pressure through a semi-permeable membrane. Water passes the membrane, while the minerals stay in the concentrate.

The solid phase is composted or granulated and used as fertilizer. The water that passes the membrane is called permeate or effluent. This permeate is supposed to be >99% free of pharmaceuticals. For this reason some water boards allow direct discharge to surface water, but this is not common practise. In some cases, the possibility to discharge to surface water has already been shown to cause problems (<http://www.gelderlander.nl/algemeen/binnenland/meer-overtredingen-bij-mestverwerker-aquapurga-1.4736759>). Other water boards only allow discharge to the sewer (Van der Hulst, 2014)(see also paragraph 3.2). Recently, a number of water boards have voiced their concern related to the pharmaceuticals and other micropollutants in the discharge of these factories to the ministry of IenM (<http://www.aanmaas.nl/nieuws/2015/10/uniforme-aanpak-van-lozingsvergunningen-afvalwater-mestverwerkingsinstallaties-noodzakelijk.html>).

The mineral concentrate can replace chemical fertilizer. For this an official recognition as fertilizer is necessary, which is still pending (Van der Hulst, 2015). This recognition is awarded if the concentrate is produced at one of the pilot plants.

The effects of manure treatment on the nutrient content of the manure has been investigated intensively in the period 2009-2011 in the project 'Pilots mineral concentrate' (www.mestverwerken.wur.nl). A more recent

update of this topic can be found in Velthof (2015). The behaviour of pharmaceuticals in this process has been reported by Lahr et al. (2014). Calculations show that part of the pharmaceuticals already disappear during the separation into a liquid and a solid phase. The nature of this disappearance is not known. Most of the antibiotics (tetracyclines and quinolones) end up in the solid phase. A small part is detected in the mineral concentrate, while in the effluent usually none of these antibiotics are found. More soluble antibiotics, such as sulfadiazine, behave differently and mostly end up in the concentrate.

Table 3 Proven techniques for treatment of manure (from www.mestverwerken.wur.nl). The load refers to the potential impact emission of pharmaceuticals to the environment.

Process	Goal	Products	Pharmaceuticals?	Load	Para-graph
Manure separation in a liquid phase and solid phase	Lower costs for transportation and/or further treatment	Solid phase: compost or granulates. Liquid phase is further treated	Most end up in solid phase, soluble ones in liquid phase	If used as alternative to raw manure: not clear. If used as alternative to chemical fertilizer: increase	3.1.2
Treat the liquid phase with ultrafiltration or reversed osmosis	Produce a mineral concentrate (to replace fertilizer)	Mineral concentrate and permeate (effluent)	Soluble ones found in mineral concentrate. In effluent usually no pharmaceuticals	If used as alternative to raw manure: decrease. If used as alternative to chemical fertilizer: increase	3.1.2
Aerate liquid phase	Remove nitrogen from liquid phase by nitrification and denitrification; remove phosphor (optional).	Water with low nitrogen (and optional low phosphor) content for watering or fertilizing	Not clear. Some degradation might be expected.	If used to water or as alternative to chemical fertilizer: increase	3.1.3
Concentrate the liquid fraction by evaporation	Reduce volume; produce a nitrogen rich concentrate (to replace chemical fertilizer)	N-concentrate, effluent, K-rich rest fraction	In K-rich rest fraction	If used as alternative to chemical fertilizer: nihil from N-concentrate, increase K-rich rest fraction	3.1.4
Digestion / co-digestion	Production of biogas for energy and heating	Biogas, digested manure (digestate)	Not clear	Not clear	3.1.5
Co-digest, separate, evaporate and granulate	Produce biogas for energy and heating, produce granules (to replace fertilizer)	Biogas, dried granulates	Not clear	Not clear	3.1.5

Table 3 CONTINUED Proven techniques for treatment of manure (from www.mestverwerken.wur.nl). The load refers to the potential impact on emissions of pharmaceuticals to the environment.

Process	Goal	Products	Pharmaceuticals?	Load	Para-graph
Compost	Produce a stabile soil enhancer, lower transport costs	Compost	Not clear	Not clear	3.1.6
Dry and granulate	Lower transport costs; produce granules (to replace fertilizer)	Dried granulates	Not clear	Not clear	-
Mix with other fertilizers and/or additives	Improve sales possibilities	Manure with additives	No influence or diluted	Equal or decreased	-
Disinfection	Obligatory when exported	Disinfected manure or granulates	Not clear. Partly broken down due to high temperature?	Not clear. Decreased?	-
Incineration	Reduce volume while recovering energy	Heat	Removed completely	Nihil	-

With respect to the emission of pharmaceuticals to the environment, it is important whether the solid phase and/or mineral concentrate are used to replace ordinary manure, or to replace chemical fertilizer. In the first case the load will probably decrease, since a part of the pharmaceuticals seems to be removed in the treatment process (Lahr et al., 2014). When however, they are used to replace chemical fertilizer (which contains no remains of pharmaceuticals), this will lead to an increased emission of pharmaceuticals to the environment.

3.1.3 *Aeration of liquid phase*

This treatment aims to remove nitrogen by nitrification and subsequent denitrification. It is also possible to remove phosphorus. The result is an effluent with low nitrogen and phosphorus content that can be used to water the land, or used as a fertilizer (depending on the remaining nutrients), or discharged via the sewer. The process is currently mainly used for calf manure.

After separation the liquid phase is aerated. The oxygen in the air converts the ammonium (NH_3) to nitrate (NO_3). In the next step the nitrate is converted to nitrogen gas (N_2). Depending on the quality of the manure it can be necessary to add a carbon source. The sludge is allowed to settle, resulting in a solid phase (20-25% of the initial volume) and an effluent (75-80% of the initial volume). It is also possible to remove phosphorus by precipitation. In this way most phosphorus remains in the solid phase. This solid phase is dried, burned or used on the land.

It is not clear what happens with pharmaceuticals during nitrification, denitrification and precipitation. Some removal might be expected, but part of the pharmaceuticals will remain in the water. When used to water crops, or when used to replace chemical fertilizer, this may either increase the emission of pharmaceuticals to the environment or change their routing.

3.1.4 *Concentrate liquid phase by aeration*

In this treatment a nitrogen rich concentrate is produced from the liquid phase. The liquid fraction is vaporized by a technique called mechanical vapour recompression. The ammonia/water vapor that is formed is washed with sulphuric acid and condensed. What remains is an N-concentrate, an effluent and a potassium rich rest fraction. The N-concentrate is used as replacement for chemical fertilizer. The effluent can be discharged or used to water agricultural land. The potassium rich rest fraction will be used on the land, most likely to replace chemical fertilizer (www.mestverwerken.wur.nl).

Generally, pharmaceuticals are not volatile. Due to the vaporization step the N-concentrate and the effluent are not expected to contain pharmaceuticals. Pharmaceuticals will remain in the potassium rich rest fraction. When this fraction is used as replacement of chemical fertilizer, this will increase the emission of pharmaceuticals to the environment or change their routing.

3.1.5 Co-digestion

Production of biogas by digesting of organic matter is a process that already has been applied for a long period of time in the Netherlands. In the period 1970-1990 this was limited mostly to small installations in which only manure was digested. The last decade the number of digestion installations has grown fast. The new generation digesters are much bigger, and in most cases manure is digested together with other biomass of plant or animal origin. This is called co-digestion, where at least 50% manure must be added to the mixture. The purpose of adding other biomass to the manure is to improve the production and the quality of the biogas, *i.e.* improve the percentage of methane and lower the H₂S production (Ehlert et al., 2013b). It is also possible to digest biomass without addition of manure, but this is beyond the scope of this report.

There are limitations to the type of organic materials that can be used for co-digestion: only organic materials that are listed in Annex Aa, part IV of the 'Uitvoeringsregeling Meststoffenwet' ('Fertilizer Law Implementation Regulation') are allowed. The products listed under category G of this Annex have to be checked by the producer for compliance with maximum allowed concentrations of metals and certain organic substances in the product, whereas all the other product categories have already been approved by applying a standardized assessment protocol⁸ which may also have included other contaminants. In order to stimulate the availability and use of organic materials as co-digestion material, studies have been performed as a basis for the addition of more products in Annex Aa, part IV (Ehlert et al., 2013b; De Poorter et al., 2013). For example, this has resulted in less strict criteria for specific pesticide residues in the organic materials, which were introduced in the standardized assessment protocol (Commissie Deskundigen Meststoffenwet (2013)).

Products of co-digestion are biogas and digestate. Biogas can be used to generate energy and heat (*i.e.* co-generation). It is also possible to upgrade the biogas to natural gas quality and deliver it back to the natural gas circuit.

The digestate is considered to be manure since it is compulsory produced with fractions of animal excrements, supplemented with one or more of the permitted products from Annex Aa. Because of this, the rules for use, transport, trading and storage of manure apply.

Cattle farms with a digester can usually apply the digestate on their own fields. Pig farms usually do not have sufficient agricultural area. In those cases the digestate is sold to other agricultural businesses, or (after disinfection) transported to foreign countries (Van Bruggen, 2012).

It is not clear what happens with the pharmaceuticals during the co-digestion process. First of all, the manure is diluted by adding organic

⁷ <http://wetten.overheid.nl/BWBR0018989/bijlageAa/>

⁸ The assessment is based on an input assessment, meaning the assessment is based on the quality of the co-digested material prior to digestion (Commissie Deskundigen Meststoffenwet, 2013).

material, thus lowering the concentration (assuming the organic material does not contain pharmaceuticals). During co-digestion, part of the pharmaceuticals will probably degrade. But when the organic matter is degraded more easily than the pharmaceuticals this might lead to concentration of pharmaceuticals in the digestate. However, relative to the amount of nitrogen, the amount of pharmaceuticals stays the same. The resulting outcome of these processes has not yet been studied.

The above-mentioned conclusion is in line with a recent report of the Dutch Soil Protection Technical Committee (TCB; 2014). TCB investigated the potential soil pollution of heavy metals and micropollutants, including (veterinary) drugs and hormones, due to the use of co-digestion digestate as fertilizer. The main conclusion is that these substances may indeed cause environmental problems, but quantitative estimates on their fate during the co-digestion process and subsequent exposure are lacking.

RIVM recently investigated the role of human pathogens in the co-digestion process related to potential environmental health problems (Van Heezen et al. 2015). It is known that several human pathogens may occur in manure, *e.g.*, various types of bacteria, viruses and parasites. RIVM concludes, however, that there are currently no indications that the distribution of human pathogens via co-digestion digestate is increased in comparison with the conventional application of untreated manure on land. A similar conclusion is drawn for the distribution of antimicrobial resistant bacteria.

3.1.6 *Compost*

Dry manure types, or the solid phase of manure (after separation), can be composted. This can be done either extensively (outside, for several months) or intensively (in an installation, for a few weeks). The composting process depends on the quality of the starting material, *i.e.* structure, carbon/nitrogen rate and moisture content. In some cases other organic matter has to be added to improve the structure.

When the compost is even produced with only a small fraction of animal excrements, the rules for use, transport, trading and storage of manure apply. When less than half of the initial material is manure, the resulting compost can be used outside agricultural areas, in which case the rules for organic fertilizers apply. When the compost is exported outside the Netherlands it has to be disinfected by heating for at least an hour at 70°C.

It is not clear what happens with pharmaceuticals in the manure during the composting process. When the organic matter breaks down quicker than the pharmaceuticals, concentration may take place. On the other hand, during the composting process high temperatures may occur, which might accelerate degradation of pharmaceuticals. These processes have not been measured however. For compost, other quality requirements may apply than for other manure-related products.

3.2 Treatment of manure together with wastewater

Several Dutch water boards see possibilities to regain valuable products such as struvite, ammonium and energy by the combined treatment of manure and wastewater (Burgers, 2012). This has synergy with the transition of the STP as a factory for resources and energy (see paragraph 2.3). It is emphasised that this technology has not yet been implemented at full-scale in the Netherlands. However, by introducing manure into the STP the emission route of veterinary pharmaceuticals to the aquatic environment changes considerably, from the indirect route via soil to the much more direct route via wastewater. For this reason this subject is included in this report.

In STOWA (2011c) an inventory has been made of the possible synergy between manure treatment and wastewater treatment. Although five different options were considered it was concluded that only two would be economically feasible. These were treatment of the permeate (*i.e.* effluent) of the reverse osmosis from the production of mineral concentrate (see paragraph 3.1.2), and separate digestion of raw manure and sewage sludge with or without combined treatment of the rejection water. From model calculations for three STPs it was concluded that - under the current market circumstances - the treatment of permeate of the reverse osmosis on a STP with available capacity offers good financial synergy. The digestion of raw manure at the STP site offers synergy with respect to the energy production and recovery of nutrients. However, with the current prices this is not (yet) financial attractive due to:

- high costs of nitrogen removal and/or recovery
- transport costs to the STP
- sales and marketing costs of the products

When nitrogen removal or recovery is not necessary to meet effluent criteria for nitrogen, the financial side is more balanced.

A project that is worth mentioning is the SOURCE project, a pilot project in which the combined treatment of humane urine with the liquid fraction of manure was investigated. The pilot was finished in 2011 (Mulder et al., 2011; Mulder et al., 2012).

The SOURCE-pilot included three major steps: 1) pre-aeration of the mixture of urine and manure to remove organic substances, 2) recovery of phosphate by struvite precipitation and 3) removal of nitrogen by a two reactor nitrification-anammox process. The effluent was discharged to the STP (Mulder, 2012). The process worked well with respect to the removal of nutrients, although the urine-manure mixture had to be diluted two or three times for stable operation. It was estimated that the costs for treatment would be comparable to other forms of manure treatment (Mulder et al., 2011; Mulder et al. 2012).

The presence of antibiotics (as the combined activity of all antibiotics belonging to a certain class) in the influent and effluent of this pilot was screened two times during the project. The activity of several classes of antibiotics was detected in the urine-manure mixture as well as in the urine and manure. During the SOURCE treatment the activity in the water phase (*e.g.*, effluent of the SOURCE pilot installation) decreased

significantly. It was not investigated whether this was due to adsorption to sludge or due to biological degradation (Mulder et al., 2012).

When manure, the liquid phase and/or permeate is treated at the STP, the STP is loaded with an additional amount of pharmaceuticals. In the case of permeate this extra amount is low, probably negligible, although this has not thoroughly been investigated. In the case of manure or the liquid phase it can be assumed that the load of pharmaceuticals in the effluent of the STP will increase. Mass balance calculations will be necessary to show whether this increase is negligible or not. The effects of antibiotics from the manure on the microbial biomass in the STP (and thus, on the functioning of the STP) would also need further investigation.

3.3 Integral concept - ECOFERM

A new integral concept to deal with waste products of intensive animal farming has been introduced: ECOFERM⁹. In this concept the waste products, *i.e.* manure, ammonia, CO₂ and heat, are used to produce duckweed, algae, biogas, electricity, heat and clean water.

In 2013 the first ECOFERM stable has been built for 1600 calves. The manure from the calves is removed several times a day. It is separated in urine and a solid and liquid phase. The solid phase is digested, producing biogas. This biogas is used to produce electricity and heat for the farm and another 140-200 households. The digested manure is dried and granulated and sold as fertilizer.

The urine and liquid phase are transported to a greenhouse that has been built on top of the stable. In this greenhouse duckweed is grown, using the minerals from the manure and the CO₂ and heat from the animals. The CO₂ promotes the growth of the duckweed. The effluent from the duckweed cultivation still contains sufficient minerals to grow algae. The duckweed and algae are fed to the calves. The algae water is claimed to be health promoting, hence lowering the need to use antibiotics.

The ECOFERM concept is aimed to close the mineral cycle and to reduce waste, CO₂ release and the purchase of food, while at the same time promoting the health of the calves. It has not been investigated what happens with pharmaceuticals in the system. It can be expected that during the cultivation of duckweed and algae, part of the pharmaceuticals degrade and part will be taken up in the duckweed and algae. If pharmaceuticals are administered to the calves it can be reasoned out that to some unknown extent a part of the pharmaceuticals will be recycled in the system. The dried and granulated digested manure, potentially containing pharmaceuticals, is exported outside the Netherlands.

⁹ <http://www.innovatienetwerk.org/nl/concepten/view/109/ECOFERM.html>
http://www.innovatienetwerk.org/~bijlagen/video/duurzaam_ondernemen/Ecoferm_eindversie_NL_OT.mp4

3.4 Aquaculture

Fish breeding for consumption

In the Netherlands fish is bred on land, using a completely closed system in which the water is constantly recirculated and treated with a biological filter system (Van Eijk, 2008; www.pvis.nl). This way hardly any water is discharged.

The production of fish for human consumption in the Netherlands was about 10,000 tons in 2008, mainly catfish (4,200 tons), eel (3,000 tons), tilapia (1,500 tons) and 1,300 tons of other species. This makes the Netherlands a very small player in aquaculture, compared to a global yearly production of 50 million tons, for 90% originating from Asia (Van Eijk, 2008). The number of sites in the Netherlands is not known.

The Dutch sector for breeding of fish in 2005 established a Code of conduct ('Gedragscode' in Dutch) for the breeders of consumption fish in the Netherlands (PVIS, 2005). With respect to veterinary pharmaceuticals, the code of conduct states that the breeder has to take proper precaution measures to prevent the outbreak of diseases. Nevertheless, if there is an outbreak, there are seven preferred substances to intervene. Furthermore the use of pharmaceuticals needs to be administrated and a minimum period of time between administration of pharmaceuticals and the consumption of fish has to be taken into account. After use of therapeutic substances, the water is not directly discharged to the surface water, unless it has been sufficiently filtered prior to discharge. In a monitoring campaign Schrap and colleagues (2003) did not find any pharmaceuticals in the effluent of sites where consumption fish were bred.

Although there is uncertainty about the increase or decrease of the production, the emission of veterinary pharmaceuticals from the breeding of fish for consumption in the Netherlands is expected to be - and stay - very limited.

Ornamental fish trade

Ornamental fish are often tropical fish. Mostly they are imported from the tropics, acclimatized and then further traded (Rijs et al., 2003). This practice makes the fish very susceptible to diseases, hence the use of antibiotics is high. The wastewater is discharged to the sewer. In a monitoring campaign of Schrap and colleagues (2003) high concentrations of antibiotics were measured in the wastewater of decorative fish breeders. More recent monitoring data could not be found however.

There are no easily accessible figures about the number of ornamental fish breeders in the Netherlands. The number is estimated to be several tens, and seems to be decreasing, based on information in the Dutch Trading Register (www.kvk.nl).

Because the concentrations are suspected to be high, more insight into the loads of pharmaceuticals resulting from ornamental fish breeders is desirable.

4 General conclusions and recommendations

4.1 Conclusions

The transition towards a circular economy, *i.e.* moving 'from waste to resource', is a very positive and promising development with many possible gains from both economic, environmental health and sustainability (the reduction of greenhouse gas emissions) perspectives. However, besides the many advantageous aspects, these new process technologies and waste application routes may also have several points of attention. Waste streams containing micropollutants may be withdrawn from conventional treatment systems and may thus enter the environment in higher amounts than before or with other emission routes than before. This may have an impact on human health (*e.g.*, via drinking water) and ecosystems. A one-dimensional approach is therefore not suitable when creating facilities towards a sound circular economy. Solving one problem should not evoke new ones.

The scope of this report was to give a brief overview of the new technological developments in waste treatment and reuse, focussing on waste containing human and veterinary pharmaceuticals. An identification was given on the 'status' of the technology, *i.e.* a distinction is made between the exploratory (lab scale or pilot plant) or operational stage. Additionally, an assessment was made on the potential influence of those processes on the release of (veterinary) pharmaceuticals into the environment.

When analysing the new technologies in many cases quantitative data on the actual occurrence or mass balances of human and veterinary pharmaceuticals was found to be lacking. This implies that judgements on possible changes of environmental loads are mostly based on expert judgement and are 'only' qualitatively scored as potential increase, potential decrease or no change. There may be several reasons why such quantitative information is not (yet) available. In pilot resource recovery projects the initial focus is on optimizing the technology rather than on analysing the presence of potential residues of micropollutants. Furthermore the chemical analysis of pharmaceuticals and their metabolites in various environmental matrices is known to be technically complicated.

In this study we focussed on potential changes in the environmental *load* of human and veterinary pharmaceuticals due to new technologies. It is emphasised that environmental emissions are not directly connected to *actual risks* of these chemicals to either man or ecosystems. However, an alteration of exposure may affect the potential risks. In addition, the possible occurrence of residues of pharmaceuticals in end products is a delicate issue from a marketing and social perception point of view.

Taken into account the above-mentioned points we made an initial overview of many projects being initiated in the Netherlands dealing with waste streams that potentially contain human and veterinary

pharmaceuticals (see Tables 1, 2 and 3). Initiatives such as post treatment of STP effluent or separate treatment of hospital wastewater are definitely lowering the emission of pharmaceuticals to the environment. Many other initiatives are inspired by the possibilities to recycle nutrients, produce biogas and/or recover resources for new products, but are not specifically aimed at the removal of micropollutants such as pharmaceuticals. Because of this, these new technologies may inadvertently increase the load of pharmaceuticals into the environment, or at least change the way pharmaceuticals are released into the environment.

From an environmental quality perspective the following technologies could be of concern, because of either possible increases of pharmaceutical emissions into the environment or unfavourable changes in their exposure routes:

- products recovered from the STP (see paragraph 2.3.3 - 2.3.7),
- digestion of sewage sludge (paragraph 2.3.2),
- use of concentrates to replace chemical fertilizer (paragraph 3.1.2 - 3.1.4),
- co-digestion or composting of manure (paragraph 3.1.5 and 3.1.6)
- treatment of manure together with wastewater at the STP (paragraph 3.2)

In paragraph 4.2 some recommendations are presented to reduce the uncertainties on the actual occurrence of human and veterinary pharmaceuticals in or from these processes.

In Chapter 1 (Figures 1 and 2) it was explained which exposure routes are currently taken into account when performing an environmental risk assessment (ERA) at the authorisation stage of (veterinary) drugs. The current study indicates that many resource recovery and sanitation technologies are still in an experimental stage. On top of that it is shown that there are several quantitative uncertainties in the environmental fate of pharmaceuticals in these technologies. Although new technologies may definitely influence, both positively and negatively, the environmental load and potential risk of human and veterinary pharmaceuticals, it is concluded that it seems too early to adapt the ERA exposure routes now already in a generic way (see also Recommendations in paragraph 4.2).

Wastewater, sewage sludge and manure contain antibiotics and antimicrobial resistance genes. Different ways of dealing with these waste streams may have an impact on the spread of antimicrobial resistance genes. As mentioned in paragraph 3.1.5, it was concluded by RIVM (Van Heezen et al. 2015) that there are currently no indications that the distribution of antimicrobial resistant genes via co-digestion digestate is increased in comparison with the conventional application routes of untreated manure on land. According to RIVM the same conclusion holds for the spread of pathogens via co-digestion digestate.

This report was focussed on pharmaceuticals consisting of organic molecules, which is the majority of pharmaceuticals. However, these molecules may behave in various ways in the environment and during treatment. Some pharmaceuticals may be degraded completely, others

are degraded to a lesser extent. Besides this, waste streams may also contain pharmaceuticals that are metals (like zinc or copper) or other persistent substances, which do not biodegrade at all. This may be a concern, since these compounds might concentrate throughout treatment of waste streams, for example during digestion or composting, and thus enter the environment in higher amounts. Only when the amount applied onto land is coupled to the amount of nutrients this concentration may be of less concern since the nutrients are then also concentrated.

The waste streams that are considered in the report (*i.e.* urine, faeces, communal wastewater, sewage sludge and manure) will not only contain pharmaceuticals but also many other micropollutants, for example biocides, cleaning agents, microplastics or personal care products. Many remarks that are made on pharmaceuticals may also apply to these micropollutants. They are however not considered in further detail in this report.

4.2 Recommendations

In paragraph 4.1 the uncertainties on the impact of new sanitation and resource recovery processes on the environmental emissions of human and veterinary pharmaceuticals were discussed. This resulted in the conclusion that it seems too early to modify the assessment schemes in the environmental risk assessment (ERA) for human and veterinary pharmaceuticals. RIVM recommends, however, to further address the topic at an EU level and emphasises that both new sanitation concepts and circular economy applications will eventually influence the methodology for assessing environmental risks of pharmaceuticals. If sufficient and reliable data are already available, some of these new insights could already be applied in the ERA, at least in a qualitative way. The ERA guidance could be made flexible with respect to exposure routes of pharmaceuticals, and keep pace with the technological developments.

Because of the lack of information on environmental fate and mass balance of pharmaceuticals in the various waste and production streams (see paragraph 4.1), RIVM recommends to further elaborate on the following research questions:

1. What is the uptake of pharmaceuticals (and other micropollutants) in products recovered from the STP?
2. What happens with pharmaceuticals (and other micropollutants) during anaerobic degradation and at higher temperatures? This question particularly applies to (co-)digestion of manure, digestion of sewage sludge, as well as composting of manure.

The need for more quantitative information on the fate of pharmaceuticals during (co-)digestion of manure and the subsequent application of digestate as soil fertilizer is in line with conclusions from a recent report of the Dutch Soil Protection Technical Committee (TCB, 2014; see paragraph 3.1.5). The filling of this data gap is even more important as the production of biogas from (co)digestion installations is already taken place at rather large scale in the Netherlands.

RIVM states that the further development and application of new sanitation and resource recovery technologies should be stimulated as it may significantly contribute to a safer, cleaner and more sustainable society. It is expected that several circular economy technologies that are now still at an experimental or pilot stage will be further optimized, and may also result in lowering the environmental emissions of pharmaceuticals.

One of the key challenges towards a sound circular economy, however, is finding the right balance between safety and sustainability aspects, especially in relation to existing legislation on waste and chemical risk management. How to weigh, for example, the reduction of greenhouse gas emissions when re-using waste streams as feedstock versus the potential presence of some hazardous chemicals in these waste stream or subsequent end products? Current legislation still hinders a smooth transition for applying and re-using waste streams for the production of energy and products. For example, some hazardous substances are not allowed any longer in the production of new products, but there may still be a legacy of these substances in waste streams. On the other hand human health and the environment should be protected without equivocation.

RIVM emphasizes, however, that more flexible and tailor-made decisions are needed in legislation. For example, the end use/application of the recycled product should be prioritised more (*e.g.*, household product versus road construction material) in decision making. Yet such decision making processes can still be rather complicated due to the underlying issues with several choices and options, each with their own set of benefits and drawbacks. Also, due to the various stakeholders involved, arriving at a joint problem perception can be a difficult task. Spijker and Van der Grinten (2014) developed a general, conceptual model which can be used as a framework to get insight and common understanding of these challenges. The practical application of this RIVM model is currently being further investigated.

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Appendix 1 Consulted persons

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- Bert Palsma, program manager water chain at the Foundation for Applied Water Research (STOWA)
- Leon de Poorter, researcher at Centre for Safety of Substances and Products (VSP) at the National Institute for Public Health and the Environment (RIVM)
- Jan-Evert van Veldhoven, senior wastewater engineer (innovation) at Waterschap de Dommel
- Nico Wortel, senior wastewater engineer at Pharmafilter
- Kirsten Zagt, manager / owner of Bareau Duurzame Technologie

