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The International Fertiliser Society

# NON-METALLIC CONTAMINANTS IN DOMESTIC WASTE, WASTEWATER AND MANURES: CONSTRAINTS TO AGRICULTURAL USE

by

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1. **ABSTRACT.**

The existing handling of biodegradable waste, wastewater and some animal manure is not organised in a sustainable manner that includes the reuse of the constituent nutrients. As our natural resources of phosphorus and potassium are finite and there is a high energy input (based mostly on fossil energy) for production of nitrogen fertilisers, a rethinking of the nutrient management procedure is essential. However recycling returns not only the nutrients but also potentially hazardous substances such as pathogens and organic pollutants to agricultural fields.

This work provides a review of current knowledge on the loads and types of pollutants to be dealt with in biodegradable waste, the different wastewater streams and manures, and describes the potential hazards, such as antibiotic resistance, contamination and uptake in agricultural plants, effects on soil-dwelling organisms, and ecotoxicological impacts. It also describes the treatment options available to reduce these compounds to an acceptable level, e.g. anaerobic digestion, composting, membrane technologies and oxidation processes. There are three options regarding the handling of these potential fertiliser sources: to continue with business as usual; to head towards a zero emissions policy that would lead to a large increase in incineration and other high-tech solutions; or to re-use these nutrient streams while taking into account the potential hazards included and addressing them by defining appropriate treatment techniques. The main non-metallic pollutants that need to be dealt with are pathogenic microorganisms. Even though organic pollutants have to be taken into account e.g. antibiotic residues can lead to major problems with resistance developing in soils. The major conclusion is that treated wastes should be regarded as a valuable fertiliser. This involves using them to replace mineral fertiliser in appropriate amounts. It is easier to handle the different pollutants separately at source rather than mixing the different fractions before treatment. Thus by source-separating into yellow, brown, and grey water, it is easier to recycle nutrients and to sanitise the different wastewater fractions according to their specific composition.



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**Key words**: contaminants, manure, organic fertiliser, organic pollutants, pathogens, treatment option, urine, faeces, waste, wastewater.

## 2. INTRODUCTION.

To maintain agricultural yields at high levels over the years, fertilisers are required. Around 50% of the fertilisers used in agriculture today are of organic origin (Mosier, 2001). As phosphorus and potassium in their present forms are finite resources and the energy and hydrogen inputs needed for producing nitrogen fertilisers are high and currently mainly covered by fossil gas, fertiliser production relies heavily on exhaustible raw materials. For long-term sustainable agriculture, the consumption of virgin nutrients has to be optimised by limiting their use, which would also minimise nutrient losses to water bodies and reduce the volume of mineral fertiliser inputs. The major flows of nutrients in agriculture are internal, firstly in the field itself and secondly, in the case of livestock farming in combination with plant production at farm level through manure management. However, the removal of crop and animal products from farms (Figure 1) removes with it large amounts of nutrients which have to be replaced. The most common way to do this is by adding mineral fertilisers. Another option is to return the nutrients to the fields as organic fertiliser originating from solid biodegradable household waste and different wastewater fractions. For developing countries this is a very relevant issue, as the need for fertilisers is extremely high while the economic potential to import mineral fertilisers at the same price as in the industrialised world is low.

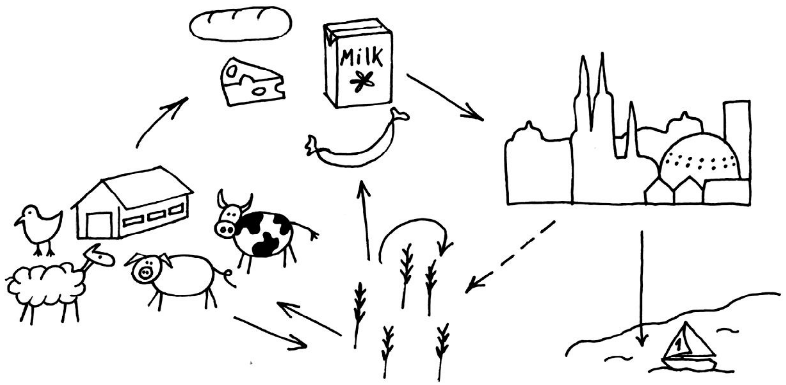


**Figure 1**: *Flows of plant nutrients in society today*.

Depending on the wealth of the social group, the quality of human nutrition is changing. In developing countries, the consumption of animal products is rather low. Compared with Western Europe, therefore, fewer animals are required to match the demand for animal products by the population and thus less animal waste is produced. For example, in the province of CanTho in Vietnam, about the same amounts of the macronutrients nitrogen and phosphorus are found in human excreta as in animal manure (Arnold and Clemens, 2004). In cases where food production is mainly in vegetable form, the percentage losses of nutrients from the farm are actually higher than on



animal farms, as a larger proportion of the harvest is exported from the farm. This is the case in most developing countries and as they cannot afford to compensate for the loss of nutrients with mineral fertiliser, there is an annual net loss of nutrients from the fields. In some parts of eastern Africa the loss can be as high as 60 kg of nitrogen per hectare and year. Locally available sources of nutrients for fertilisation are actually only domestic waste and wastewater. Today, these fractions are in most cases not utilised but rather regarded as pollutants in the environment, destroying recipient waters, and any utilisation that does occur takes place in an uncontrolled and unsafe way.

Together with world-wide development and improved living conditions, demand and consumption of animal products is increasing. To meet this demand, animal production is also increasing. However, increased animal production requires increased feed production and this increases the need for plant-available nutrients at farm level.

There is a growing tendency for urbanisation and during 2007 the number of people living in cities exceeded the number of people living in rural areas. According to the UN, more than two-thirds of the population will live in cities in 2050, while the world's population will have grown to approximately 9 billion. Nutrient flows to cities will increase in the form of agricultural products imported and there will be a concomitant increase in waste and wastewater flows.

Animal waste is already an important source of plant nutrients in agriculture. For example, in Germany the amounts of nitrogen and phosphorus in manure that are recycled to agriculture are in the same range as those in mineral fertilisers. To a certain extent, animal manure management is rather well developed, although there are still high losses of ammonia during storage and field application. However, the overall goal is to minimise losses and to maximise fertiliser use. In contrast, the current product of human wastewater treatment (sewage sludge) has a rather low concentration of nutrients. In addition, the overall process of waste treatment aims at nutrient elimination and not at nutrient recycling. By introducing source-separation of waste and wastewater systems, new possibilities are created to collect smaller fractions with relatively concentrated nutrients (Vinnerås *et al*., 2006) that can be reused in agriculture with available equipment. If all the nutrients in human excreta were collected and reused, approximately 20% of the mineral fertiliser used in the respective country could be replaced, with some variation depending on country and element. For developing countries the contribution to national consumption of mineral fertilisers would be larger, especially south of Saharan Africa where excreta correspond to more than 100% of the local consumption of mineral fertilisers (Rockström *et al*., 2005).

### 2.1. Domestic waste, wastewater and manures.

Domestic waste and industrial organic waste from restaurants and kitchens is a large fraction that is easily collected separately. The composition of the waste is related to the time of the year, as food consumption follows the seasons to some extent, e.g. in Northern Europe citrus fruit peel is found to a higher extent during autumn and winter compared with spring and summer. Parts of these fractions are reused, either as fertilisers or as soil conditioners, while others are incinerated or land-filled, depending on the local setting. Before reuse as fertilisers or soil conditioners most of these materials are either composted or anaerobically digested. Some biodegradable industrial waste can be applied directly without treatment, e.g. mycelia from pharmaceutical industries, but in most cases it is biologically stabilised before reuse when storage is needed due to seasonal factors.

Household wastewater can be divided into separate fractions according to their source. This separation allows separate management of the fractions as they are very different in composition due to their origin. The main water fraction consists of greywater (all household water fractions except toilet water, including wastewater from sinks, showers, washing, etc.) and this fraction contains the majority of the metallic pollutants and only a minority of the nutrients (Figure 2). An exception has to be made where high amounts of phosphorus detergents are used, especially for dishwashers and laundry, but greywater is generally not of any major interest in a nutrient perspective. The main reason for greywater reuse is water shortage, when this wastewater stream can be looked upon as an additional water source. Another interest in greywater reuse is to decrease the household environmental impact or as a combination of the two. Greywater can either be used locally in households, after some treatment, for non-potable usage e.g. flushing toilets, or for other purposes such as irrigation, car washing, cleaning streets, etc.

The content of organic pollutants in greywater, like the content of non-organic substances present, is a reflection of the substances used in households. The main concern arising from greywater is its content of organic pollutants and pathogens. Substances such as pharmaceuticals, pesticides, fragrances and flame-retardants can be present in greywater. Eriksson *et al*. (2003) identified more than 190 organic compounds belonging to the surfactants, emulsifiers, fragrances and flavours, preservatives and antioxidants, softeners and plasticisers, UV-filters, solvents, dyes and others in Danish greywater entering the wastewater stream due to ongoing household activities, i.e. personal hygiene and cleaning. Studies by Palmquist and Hanaeus (2005) also showed the presence of the antibiotic triclosan at notable levels (0.6-6 $\mu g/L$).

In comparison with other fractions, the level of pathogens in greywater is low and mainly based on faecal contamination, which has been estimated to correspond to 40 mg / person / day (Ottoson and Stenström, 2003). Even with this low grade of contamination, Ottoson and Stenström (2003) reported a risk of transmission of viral diseases, using the example of Rotavirus. They also reported growth of pathogenic bacteria within the system, e.g. *Salmonella* spp. and *Escherichia coli* O157, as the concentration of the faecal indicator bacteria *E. coli* in the system was considerably higher than the measured contamination entering the system. This was confirmed by studies performed by Friedler et al. (2006) and Winward et al. (2008) who found an average level of faecal coliforms of over $10^5$ colony forming units (cfu), $ml^{-1}$, which indicates



growth within the system. This has to be taken into account when selecting the mode of application of greywater.

The two excreta fractions urine (yellow water) and faeces (brown water) can be collected separately from each other or together as blackwater / toiletwater. As can be seen in Figure 2, the main contribution of nutrients originates from urine, which contains 90% of the nitrogen and 70% of phosphorus and potassium in excreta (Vinnerås *et al*., 2006). In terms of the plant availability, urine contains even more of the nutrients due to the fact that the nutrients in urine are contained in ionic form while in faeces they are mainly bound to organic compounds and cell mass. The composition of these fractions is based on the human metabolic system, as the metabolised substances mainly end up in the urine while the non-metabolised material remains in the faeces, explaining the high nutrient content and the low heavy metal content in urine.

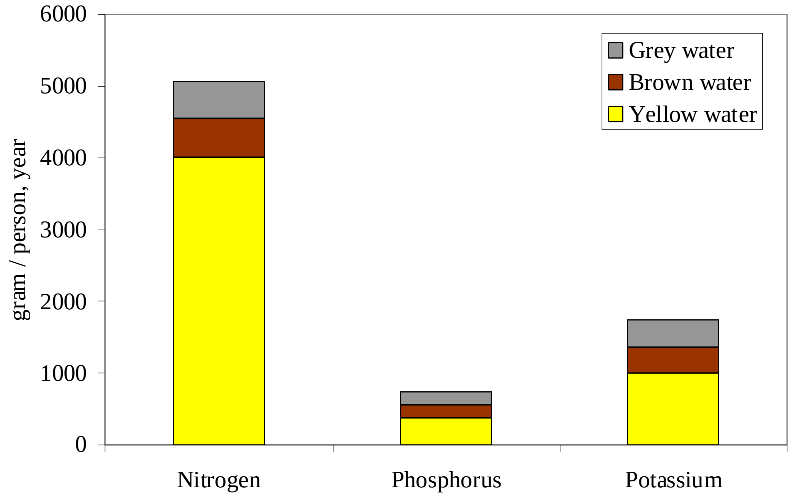


**Figure 2**: *The distribution of nutrients in biodegradable household waste and wastewater fractions in Sweden (Vinnerås et al., 2006).*

### *2.1.1. Pharmaceutical residues.*

An exception to this distribution pattern among the different wastewater streams is found for organic pollutants such as pharmaceutical residues. They are excreted in urine and faeces depending on their metabolic characteristics. The separation of organic compounds with urine is mainly on the basis of charged substances (negatively charged acids or positively charged bases) and by the liver and bile mainly on the basis of more non-polar substances.

Most statements regarding pharmaceuticals in different wastewater fractions are based on calculations from data given by the medical companies producing the substances. So far, only measurements of pharmaceutical residues in urine exist (Winker *et al*., 2008a). Figure 3 (overleaf) shows the mean concentrations recorded in some measuring campaigns in Germany

(Strompen *et al*., 2003; Tettenborn *et al*., 2007) in relation to the amount of samples taken, the detection frequency and the limit of quantification. Besides the active substances shown in Figure 3, acetylsalicylic acid, phenofibrate, gemfibrozil, indomethacone, ketoprofen as well as the hormones estriol, 17ß-estradiol, 17α-ethinylestradiol and mestranol (a pro-drug of 17α-ethinylestradiol) were investigated but remained below the limit of quantification.

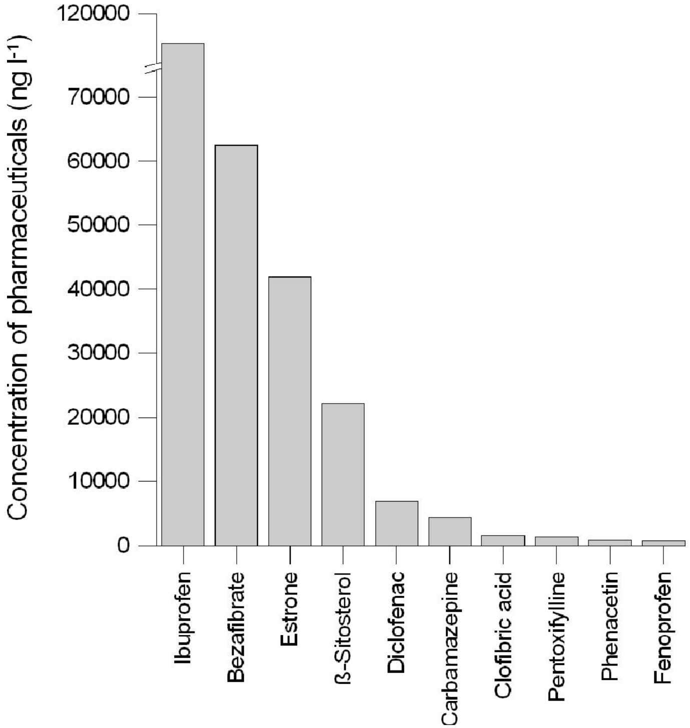


**Figure 3**: *Measured mean concentrations of active agents in German source-separated urine (Strompen et al., 2003; Tettenborn et al., 2007).*

Concentrations of pharmaceutical residues in faeces have only been estimated via theoretical calculations (Winker *et al*., 2008b). It could be shown that pharmaceuticals are excreted to a large part via urine and only partially via faeces, a fact supported by Lienert et al. (2007a) reporting on calculations that around 70% are excreted via urine. However, some substances also show high faecal excretion rates, e.g. erythromycin (50%) and terbutaline (60%) (FachInfo-Service, 2005). A large proportion of the pharmaceutical substances that are metabolised retain their activity when excreted or are easily biologically degraded to a form that has biological effects. The reduction of pharmaceuticals starts directly when the substance enters the blood stream. Therefore, the dose of the consumed substances is soon reflected in the excreta (Figure 4), as the half-life within the body of most pharmaceuticals is short.

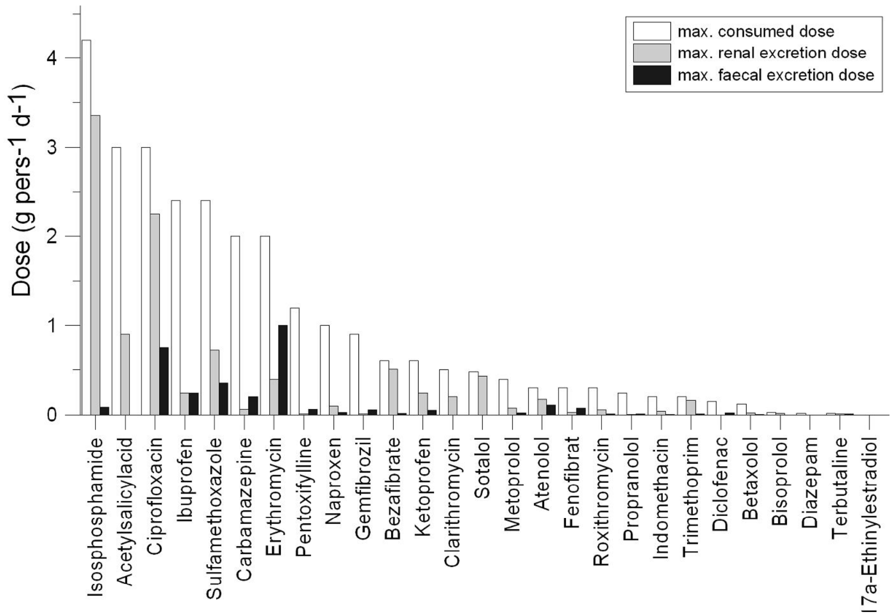


**Figure 4**: *Maximum consumed daily doses for a single patient and their related amounts excreted via urine and faeces calculated for some active substances (Winker et al., 2008a).*

Organic pollutants in solid biodegradable waste (biowaste) are mainly of two kinds. The first is misplaced medicines, a source that it is hoped can be eliminated by proper information. The second and major source of organic pollutants is substances used in agriculture for promoting the growth of the plants, e.g. pesticides and herbicides. These substances are mainly found on the surface of vegetables, which in most cases is the part removed during preparation. Therefore the levels of these substances are higher in the biowaste than in the parts of the plant that are consumed. As the substances used originally come from the field, the effect in the field from recycling these substances should be marginal compared with the effects from the fraction of the substances that does not leave the field. The major effect is rather a risk for decreased efficiency in waste treatment, as these substances may affect the microbiota within the compost or digestor.

### *2.1.2. Pathogens in urine.*

The excretion of pathogens is the converse of pharmaceutical excretion as the majority are found in the faeces and there are only a few pathogens that can be excreted via the urine. The main transmission route of pathogens in urine is actually cross-contamination from faeces during collection (Höglund, 2001).

The main pathogens found in urine are *Salmonella typhi/patatyphi*, *Mycobacterium tuberculosis*, *Leptospira interrogans,* and *Schistosoma haematobium* (Faechem *et al*., 1983). The two organisms listed first are normally not excreted in urine and appear in urine only on very rare occasions. *Salmonella typhi/paratyphi* is only excreted by people with typhoid/paratyphoid fever, where the bacteria are disseminated in the blood. *Mycobacterium tuberculosis* is also only transmitted into urine in cases of renal tuberculosis. For *Leptospira interrogans*, which is a zoonose, urine is not considered to be an important transmission route as the prevalence in human urine is low, whereas *Schistosoma haematobium* larvae are renally excreted. However, the larvae have a short survival span, that can be counted in days, during their transport after release from humans as eggs. These eggs hatch on reaching freshwater into a free-swimming miracidium that infects freshwater snails. Therefore urine reuse as fertiliser is not considered to be a major transmission route, as long as the urine is stored for at least one week.

### *2.1.3. Pathogens in faeces.*

The main risk for transmission of diseases in systems where organic waste and wastewater are recycled are pathogenic organisms excreted with the faeces. The pathogenic organisms can be divided into four categories, non-spore forming bacteria, spore forming bacteria, viruses and parasites. In addition these groups can be divided into two sub-groups, those that are specific to one species and zoonoses that can infect between animal and humans, e.g. *Salmonella* spp..

The Gram-negative rod-shaped group of pathogenic enterobacteria is one of the main disease-causing groups of organisms. In addition, several members of this group are considered to be zoonoses, which increases the risk with these pathogens within the system of recycling organic fertilisers, as the recipients can be both humans and animals. The organisms that affect people most are *Salmonella* spp.. The infection dose can be as small as 20 cells, but in most cases $10^{4-6}$ cells are required for infection. However, as for all organisms, the infection dose varies from host to host depending on several factors such as age, time of infection, nutritional status, etc. and the most severe infections occur in young and old people and immunocompromised individuals. Growth of *Salmonella* spp. has been reported in sewage sludge (Sahlström *et al*., 2004), compost material (Elving *et al*., 2008) and manured soil (Gibbs *et al*., 1997). Verotoxin-producing *Escherichia coli* (VTEC) is a species of *E. coli* that produces verotoxins, a shiga like toxin. The behaviour of the organisms is similar to that of non-verotoxin-producing *E coli*, which means that they have high survival in the environment. The infection dose of VTEC is as low as 10 organisms, depending of the receptivity of the host. *Shigella* spp. is a gram-negative rod-shaped bacteria closely related to *Escherichia coli* and *Salmonella* spp. Some of the *Shigella* strains produce enterotoxin and shiga toxin, which is similar to the verotoxin found in VTEC (e.g. *E. coli* O157:H7). *Campylobacter jujeni* is a micro-aerophilic bacteria that is sensitive to high $O_2$ levels and the bacteria are sensitive to most factors related to biological



treatment, such as heating, drying and acids. Therefore this organism should not be of great concern regarding waste and wastewater management.

*Mycobacterium tuberculosis* is an obligate aerobe acid-fast Gram-positive bacteria that causes tuberculosis. *M. tuberculosis* is not generally an intestinal infecting bacteria, and although it can colonise the intestine and the renal system, the main site of infection is the lungs. However, the organisms excreted with the mucus are most often swallowed and end up in the faeces. *M. tuberculosis* is a slow-growing type of bacteria but very hardy against chemicals and drying. It is relatively stable in the environment but is thermally inactivated, with a decimal reduction of less than one day in compost at 55°C (Grewal *et al*., 2006).

Several viruses are included in the group that cause gastroenteritis, but only few of the viruses are considered to be zoonotic, e.g. hepatitis E infecting both humans and pigs, and most of them only infect one species. Other viruses commonly included in this group are hepatitis A, rotaviruses, noroviruses, astroviruses, caliciviruses, enteric adenoviruses, enteroviruses and parvovirus. In total, more than 130 viruses may be present in faecal-contaminated material. The infection dose of viruses is generally low, even as low as one single virus. Outside the body, viruses do not multiply as they need their host for growth. Some of these viruses are very thermoresistant and studies show slow reduction under high temperatures (Sahlström et al., 2008) but treatment in thermophilic composting including mixing of the material should be sufficient for proper reduction (Faechem *et al*., 1983; Vinnerås *et al*., 2003; Wichuk and McCartney, 2007).

Parasites can be divided into two groups, protozoa and helminths. Both groups have a low infection dose but they cannot increase in numbers outside the host. The protozoa, mainly the zoonotic *Giardia* and *Chryptosporidium*, are relatively sensitive towards heat treatment and are rapidly reduced under such conditions (Faechem *et al*., 1983; Wichuk and McCartney, 2007). The helminths, on the other hand, can withstand high temperatures and are pH-tolerant microorganisms, with *Ascaris* spp. looked upon as one of the most thermoresistant and chemoresistant pathogens (Faechem, 1983; Haug 1993).

### *2.1.4. Pathogens in wastewater sludge and biodegradable solid waste.*

Wastewater sludge originates in most cases from a mixture of the household wastewater fractions described, together with industrial wastewater and stormwater. Therefore, all the above-mentioned pollutants can be found in this fraction. Studies by Sahlström *et al*. (2004) indicated that within the sewage treatment plant, *Salmonella* spp. in the in-house flora is constantly contaminating the sewage sludge. The levels of organic pollutants are very closely related to the kind of industries that are connected to the wastewater treatment plant and their prior treatment of the outgoing wastewater. This makes general predictions on the level of pollutants in sewage sludge difficult and the only overall statement possible is that the quality of the sludge is lower than that of the sorted fractions in the households.

The risk for pathogens in the biodegradable waste is low in comparison with the wastewater fractions. The main risks here are instead plant pathogens and unwanted plant seeds. These can be managed in similar ways to other animal pathogens during the treatment, see below.

### 2.1.5. Effects of organic pollutants on soil fauna.

The effects of pharmaceutical residues contained in source-separated wastewater streams are largely unknown for soil organisms. Furthermore, only limited research is available so far as regards the other organic pollutants in wastewater streams (Baguer *et al*., 2000). Organic pollutants can be expected to disrupt physiological processes and to have an influence on the reproduction rate (Kolpin *et al*., 2002), and generally to have a negative influence on soil-dwelling organisms (Boxall *et al*., 2003). Nevertheless, although urine has been used in Swedish agriculture for some years, consequences due to application of source-separated wastewater streams in agriculture are unknown although it seems that soil organisms can cope with the burden contained.

It has been proved that pharmaceuticals can hinder the growth of soil organisms, e.g. mould fungi have shorter hyphae (van Gool, 1993). Earthworms, springtails and enchytraeids, the group to which most existing research in soil refers, have been found not to be affected by antibiotics (Alexy *et al*., 2004). Nevertheless, indirect effects by changes in the microbial community could occur and in the case of antibiotics there is a risk of the development of resistant bacteria. The spread of resistance is already a very dangerous trend that is occurring within hospitals and nurseries (Bonten *et al*., 2001), but the emergence and spread of antibiotic resistance is complex and difficult to study, especially as different antibiotics are applied in agriculture and might result in novel types of resistance (Smith *et al*., 2005). The constantly growing number of cases of antibiotic-resistant bacteria in the environment and during treatment of humans and animals (Bengtsson *et al*., 2008) indicates the risks associated with uncontrolled spreading of antibiotics in the environment. This is of great importance in agriculture in particular, as the recycling loop of substances and organisms is relatively short due to the intensive usage of fields with a continuous annual production cycle. Therefore, microorganisms carrying different types of resistance can survive in the soil after small continuous additions of new antibiotics and can reinfect animals and humans.

Most existing research is related to single organic substances, often even in the laboratory. This means that no relevant practical conclusions can be drawn at present. In addition, data only exist for a few combinations of two or more organic pollutants with active agents. For example, Baguer *et al*. (2000) reported a toxicity to soil fauna of the two antibiotics oxytetracycline and tylosin ($EC_{10}$ = 150 mg/kg). Moreover, even fewer data are available on chronic toxicity, while information regarding the long-term influence and changes in soil fauna is completely lacking.



### 2.1.6. Plant uptake.

The uptake of pharmaceuticals in plants and the influences they exert on plant physiology and development are of major interest, especially for agricultural crops in relation to fertilisation using waste, wastewater products and manure. Data from the literature show that plants are generally able to take up pharmaceuticals in such a way that they can be detected in roots and in aerial plant parts (Winker and Behrendt, 2008). The concentrations detected in plant parts are in the range of ng/kg. Pharmaceuticals have also been found in edible plant parts such as carrot roots and cereal grains. Dolliver *et al*. (2007) and Boxall *et al*. (2006) reported that concentrations in potato tubers and carrot roots are higher in the peel than in the centre, and concluded that there is some penetration of pharmaceuticals through the peel. In addition, Brian *et al*. (1951) and Stokes (1954) reported excretion of griseofluvin via guttation drops at the leaf apex of wheat seedlings. The rate of movement in plants is influenced directly by rate of transpiration, which in turn is affected by air humidity and temperature. This finding leads to two apparent contradictions. On the one hand, pharmaceuticals accumulate in leaves (Brian *et al*., 1951; Stokes, 1954), and higher uptake rates have been found in older leaves (Grote *et al*., 2004). On the other hand, leaves are able to secrete pharmaceuticals (Brian *et al*., 1951; Stokes, 1954) and to degrade organic chemicals taken up, in a process comparable to liver metabolism (Komoßa *et al*., 1995). The difference to human beings is that instead of excretion, in most cases compartmentalisation occurs. This means that transformation products are stored inside the plants in lignin or cell walls.

Pharmaceutical concentrations in plants depend on amounts of pharmaceuticals available in the respective growth medium. Therefore, mapping of 'real' concentrations in plant parts is nearly impossible. Often very high doses are applied in experiments to ensure the possibility of analytical detection. Kumar *et al*. (2005) reported that the correlation between the concentration applied and uptake is nearly linear, but it is currently impossible to generalise on these findings, especially since Kumar *et al*. (2005) found differences regarding the intensity of pharmaceutical uptake among the three plant species investigated in their study.

Pharmaceuticals also cause phytotoxic effects depending on the concentration of the pharmaceutical used. A change of colour to darker green (Grote *et al*., 2004) as well as lacking and incomplete colouring have been observed (von Euler, 1948; Rosen, 1954). Moreover, lower chlorophyll content in leaves (von Euler and Stein, 1955) as well as hard and waxy leaves have been reported (Rosen, 1954). Germination itself also seems to be affected; speeding up (Barton and MaeNab, 1954) or slowing down germination (von Euler, 1948; Ritter, 2008) in certain cases. Moreover, Rosen (1954) reported a lack of lateral root development subsequent to pharmaceutical exposure and von Euler (1948) found thickened coleoptiles.

Studies have shown that different plant species have differing sensitivity levels towards the same pharmaceutical. However, it must be pointed out that many articles were published 20 to 30 years ago and the sensitivity and selectivity of chemical analyses at that time was somewhat lower. Furthermore, it is not possible to extend these conclusions to long-term effects in general, as most tests described in the literature did not last for a whole growing season.

#### *2.1.7. Ecotoxicological impact of pharmaceutical residues.*

Due to lack of data, it is currently not possible to evaluate the potential toxic effects of pharmaceuticals ingested by humans in the form of crops fertilised with waste, wastewater products or animal manures. Nevertheless, several efforts have been undertaken to extend and summarise existing knowledge. The Swedish Association of the Pharmaceutical Industry has produced an environmental classification system for pharmaceuticals (Johansson *et al*., 2004). This classification includes more than 300 pharmaceuticals and their risk is analysed with respect to the volumes consumed annually in Sweden via PEC / PNEC (Stockholms läns landsting, 2008) as well as their persistence (P), bioaccumulation (B), and toxicity (T), merged in the so-called PBT index. A significant risk in terms of the PEC / PNEC ratio was only detected for the two hormones 17ß-estradiol and 17α-ethinylestradiol, but 30 substances reached the highest level for the PBT index and pose an environmental hazard (Johansson *et al*., 2004). However, it has to be mentioned that for 16 of the 30 pharmaceuticals, data are insufficient and the assessment remains uncertain.

An ecotoxicological hazard assessment has been reported by Lienert *et al*. (2007b) for approximately 40 pharmaceuticals considering their excretion. It was found that metabolic processes in the human body reduced the toxic potential of all but eight drugs investigated. Source separation of urine could remove 50% of the toxic potential but, as already discussed, does not apply for all pharmaceuticals due to varying excretion routes. Moreover, Lienert *et al*. (2007b) were able to demonstrate that ibuprofen dominated a mixture of 30 pharmaceuticals by representing 52% of the mixture's toxicity. This is an important finding as most investigations focus on single substances and if urine is considered as a fertiliser, there will always be a mix of various substances present.

Furthermore, Escher *et al*. (2005) showed that the toxicity of source separated urine with and without pharmaceuticals towards bacteria and algae did not vary significantly. Meanwhile Muskolus (2008) investigated the overall toxic potential of urine toward soil organisms by looking at earthworms as vector organisms and concluded that application of urine on agricultural land has a positive effect on plants and microbial organisms due to the organic and mineral nutrients it contains. Nevertheless, single constituents such as ammonia are acutely toxic toward worms. Moistening of the skin of earthworms with ammonia solution is lethal within a short period of time (Muskolus, 2008). This is due to electrical conductivity in combination with pH and ammonium (Nguyen *et al*., 2008). Therefore, it is important that urine application is accompanied by active incorporation into the soil, thereby avoiding drainage into worm channels. Toxic effects on worms result in a decrease in populations, which recover only slowly in a dry summer or climate.



Overall, it can be concluded that the acute and chronic effects caused by fertilisation, as well as changes in soil-dwelling organisms, are practically unknown. Hence, further research regarding the ecotoxicological impacts of pharmaceuticals on soil ecosystems is urgently needed.

## 3. TREATMENT TECHNOLOGIES FOR PRODUCTION OF SAFE NUTRIENTS.

### 3.1. Anaerobic digestion.

Anaerobic treatment is mainly performed as a mesophilic treatment, at temperatures between 30° and 40°C, but some systems are designed for thermophilic treatment at 45-55°C. As the majority of the energy produced in the anaerobic treatment is bound as chemical energy in the form of methane, the system needs to be heated by external energy sources, although in tropical climates the temperature in mesophilic reactors can be regulated by the ambient temperature.

#### *3.1.1. Pathogen reduction.*

Monitoring of pathogen reduction in different stages of treatment of biodegradable waste including wastewater products and manure shows that conventional treatment by mesophilic digestion gives a limited reduction in the content of pathogens (Table 1).

**Table 1.** *Potential percentage reduction in pathogenic microorganisms during treatment of biodegradable waste and wastewater, including animal manure (Eisenhart et al., 1977; Gantzer et al., 2001; Gerardi 2003; Horan et al., 2004; Yen-Phi et al, 2008).*

| | **Percentage reduction** | | |
|---|---|---|---|
| | *Enteric bacterial pathogens*[1] | *Parasites*[2] | *Viruses*[3] |
| Mesophilic digestion (35°C) | 0-90 | 0-50 | 0-90 |
| Thermophilic digestion (55°C) | 90-100 | 90-100 | 90-100 |

[1] The organisms included here are mainly pathogens from the *Enterobacteriaceae* group, e.g. *Salmonella* spp. *E. coli, Campylobacter jejuni*.

[2] *Ascaris* spp. used as model parasite.

[3] Adenovirus used as model for viruses. When data are not available, other pathogenic viruses are used, e.g. Hepatitis A, Rotavirus, and for pasteurisation Parvovirus.

Most studies show that conventional mesophilic treatment can reduce the incoming organisms by up to one logarithm (90 %) during the treatment, but it may be possible to improve this either by changing the chemical composition of the substrate, e.g. increasing the concentration of uncharged ammonia in the process, or by physical alterations such as temperature peaks or an increase in the minimal retention time. Using thermophilic digestion or extending treatment by composting or storage of the treated sewage sludge improves the final hygiene quality. However, long-term storage or composting decreases the fertiliser value of the final product and increases the risk of pollution of the environment, mainly through ammonia emissions. Table 1 summaries a number of studies of the reduction of pathogens during anaerobic digestion. When considering the final reduction of indicator bacteria or bacterial pathogens, both the inactivation and the regrowth after treatment due to recontamination must be considered (Albihn and Vinnerås, 2008). The data given for bacteria are therefore uncertain, e.g. Sahlström *et al*. (2004) found evidence that indicated growth of *Salmonella* spp. within the sludge management system of sewage treatment plants, as the same genotype of *Salmonella* spp. was found in the outgoing sludge during several months of monitoring. One important conclusion is that high temperature treatment (pasteurisation) is the most effective treatment, but even during pasteurisation it is possible for some viruses to survive.

The two main factors influencing hygiene are the hydraulic retention time (HRT) and the minimal retention time (MRT). The HRT indicates the average time a substrate particle remains in the reactor. With a completely mixed reactor the HRT gives the dilution factor of the incoming material and a combination with the minimal retention time is needed to estimate the potential pathogen reduction within the process. With a HRT of >10 days, the dilution factor of the fresh material will be more or less 90-99%, equal to a $\log_{10}$ dilution of the incoming material. The minimal retention time is the shortest time possible in a system from input to output. The sanitisation within the process is then calculated based on a combination of the minimal retention, giving the reduction in the process together with the hydraulic retention time that gives the dilution factor, which normally corresponds to one decimal reduction. In non-mixed processes, e.g. plastic tube digesters, the HRT and the MRT are more closely linked and as the MRT is just slightly shorter there is never complete plug flow but some turbulence, resulting in some smaller mixing. In those systems with no turbulent flow, larger non-motile organisms such as *Ascaris* spp. sediment during the treatment process, which leads to an even longer retention time compared with the rest of the material, and therefore an improved reduction for these organisms. Studies by Yen-Phi *et al*. (2008) showed a relationship between the reduction and the retention time in a plug flow reactor. Increasing the HRT from 3 days to 30 days increased the reduction of *Salmonella* spp from approximately 1 to 3 $\log_{10}$ during the process.

In conclusion, it is impossible to have complete inactivation of pathogens in mesophilic anaerobic treatment without an external input such as pasteurisation. However, a study performed by Ottoson *et al*. (2008) shows the possibility for in situ sanitisation during mesophilic digestion, with a MRT of less than 2 days. This study utilised the effect of a high protein feed of the digestor that resulted in increased pH and raised levels of ammonia, while still continuing as a functional process.



The thermophilic system requires external heating and is thereby a complex system, including stirring and heat exchangers. Regarding the hygiene in this system, there is a good inactivation of pathogenic organisms correlated to the temperature with temperatures above 50°C. However, some thermoresistant viruses, such as *Porcine parvovirus*, can survive relatively well under these conditions (Sahlström *et al*., 2008).

#### *3.1.2. Inactivation of organic pollutants.*

The effects of anaerobic treatment on pharmaceutical residues in wastewater are similar to those on pathogens. For example, in the case of anaerobic digestion of wastewater, the degradation ranges from 0% (carbamazepine) up to 99% for sulphamethoxazole (Carballa *et al*., 2006). Even within the same indication group there are large variations amongst the different substances regarding their degradation behaviour. While ciprofloxacin and norfloxacin are degraded only to around 10% (Golet *et al*., 2003), erythromycin lies around 50% (Amin *et al*., 2006) and roxithromycin even reaches up to 85-95% (Carballa *et al*., 2006). This indicates that degradation is achieved but only for certain substances. Leven and Schnürer (2005) compared the degradation of organic compounds such as benzoic acid, phthalic acid, methylphthalate, phenol and cresol and found much more efficient degradation in the mesophilic system, at 37°C, compared to the thermophilic system, at 55°C, where only benzoic acid was mineralised. Decreasing the temperature in the thermophilic system to 48° instead of 55°C had a remarkable effect on the degradation, with increased mineralisation of the substances investigated. They attributed the difference to a lower denaturisation of proteins at the higher the temperatures. However, decreasing the treatment temperature decreases the reduction in pathogens present (see above) and thereby increases the risk of spreading diseases.

### 3.2. Composting.

#### *3.2.1. Pathogen reduction.*

In the composting process, independent of the organism type, the inactivation is mainly driven by heat inactivation. The heat stresses the organisms, with the end-result that proteins are denatured. During mild heat stress the effect on the microorganisms is a reversible inactivation. As a rule of thumb used in most situations, a temperature of at least 50-55°C is needed for successful inactivation. Generally, no pathogenic bacteria grow at these temperatures but some organisms can withstand high temperature stress for long time, at temperatures up to 50°C and sometimes above, before being inactivated.

The higher the temperature the faster and more efficient the inactivation, and the better heat transfer the faster the inactivation, i.e. moist heat is more efficient than dry heat. Thermotolerant organisms that withstand high temperatures, such as those active in the compost during the thermophilic phase, are normally not considered pathogenic. The main exception to this is some spore-forming bacteria, which in their protected spore form withstand high temperatures without inactivation. These organisms mainly affect animals and are thereby mainly found in the manure, e.g. *Clostridium* *schauvoei* which causes black leg. In addition, some organisms mainly affect farming processes as they cause the death of animals due to toxin production, e.g. *Clostridium botulinum*. The main effect of the toxins emerges when they are produced in the fodder, e.g. in silage, or in the bedding in chicken farms. In areas where these organisms are considered to be a problem, special care should be taken and additional barriers should be introduced in combination with additional treatment if the material is considered for use in agriculture at all. Otherwise it may rather be a matter of disposal. The other spore-forming organism group, *Bacillus* spp., includes the pathogen *Bacillus anthraces*. However, anthrax is not commonly associated with waste and manure and is thus beyond the scope of this paper. Spore-forming organisms in general are very heat-resistant in their spore form and will probably not be significantly reduced during composting treatment. In anaerobic treatment there is even a risk of an increase in numbers of *Clostridia* spp. On farms experiencing problems due to spore-formers, extra measures need to be taken regarding manure management to reduce the spread of these organisms within the farm and to the surrounding environment.

In both anaerobic and aerobic treatment, the degradation of the organic material decreases the risk of recontamination or regrowth of pathogenic bacteria (Sidhu *et al*., 2001), as the stabilised material has a low content of easily available carbon and the count of other non-pathogenic organisms is high. Biological treatment thereby reduces the risk of re-growth and recontamination of the treated material.

### *3.2.2. Composting and other techniques for removal of organic pollutants.*

Practically no studies have been carried out on the effect of composting on organic pollutants. A study by Guerin (2001) reported that soil composting at mesophilic temperatures successfully degraded probenecide and methaqulone. This can be seen as an interesting result but testing with other drugs is necessary before any conclusions can be drawn about this technique. Nevertheless, due to the fact that many drugs are not thermally stable, many more may be degraded at mesophilic temperature.

In the case of pharmaceutical residues, membrane technologies are a promising option. Micro- and ultra-filtration membranes have too large a pore size, so substances can still pass through (Lindner, 2008). Better elimination rates have been reported for nanofiltration (Pronk *et al*., 2006). In addition, membrane bioreactors show very good results for biodegradable substances (Quintana *et al*., 2005; Braga *et al*., 2005; Cartinella *et al*., 2006). A good option would be an additional chemical treatment, e.g. ozonation.

## 3.3. Chemical treatment for removal of pathogens.

Biodegradable waste and wastewater products can also be treated and stabilised using chemical treatment. The traditional treatment for chemical removal of pathogenic contaminants is lime. The lime increases the pH, thus inactivating the organisms. Most organisms will be reduced but some are very



resistant to high pH, especially *Ascaris* spp. Studies have shown survival for more than one week at pH >13 and the general recommendation is that the pH must be kept above 12 for over three months in order to inactivate *Ascaris* spp. (Pecson *et al*., 2007; Nordin *et al*., 2008). The effect of high pH on pharmaceuticals varies greatly depending on their sensitivity, as high pH in some cases irreversibly changes the structure, mainly due to hydrolysis.

The most efficient chemical treatment method for removal of pathogens in organic material intended for reuse in agriculture and even for management of pathogen-contaminated animal manure is ammonia treatment. The ammonia can be applied either in the form of ammonia or in the form of urea, which is degraded into ammonia in contact with naturally-occurring enzymes. Ammonia treatment only needs a pH of around 9 for efficient reduction of pathogenic microorganisms (Vinnerås, 2007; Nordin *et al*., 2008). This is the hygiene-regulating mechanism in management of human urine, as the high nitrogen concentration supports the inactivation of any pathogens present (Vinnerås *et al*., 2008). The active substance is the uncharged ammonia $NH_3$, which in viruses passes into the organism and destroys the genome. The effects in other higher organisms are not as well defined. There is a potential for using ammonia for management of pharmaceuticals, but the effect is unclear even though the presence of ammonia changes the structure of organic material and thereby also the structure of organic material such as pharmaceuticals. The advantage of urine ammonia as a treatment agent is that the ammonia is not consumed during the treatment, and can therefore be used as a fertiliser after treatment.

## 3.4. Chemical treatment for removal of organic pollutants.

One of the most investigated treatment options for pharmaceuticals in wastewater effluent is oxidation, but even though it is widely discussed as a tertiary treatment in wastewater treatment plants, large-scale implementation has not yet begun. Management of the pharmaceuticals in the solid sludge stream is seldom discussed or investigated. The oxidative treatment of liquid fractions is relatively efficient in inactivating the substances by altering their structure, mainly by attaching oxygen into different places on the organic molecules. A number of substances can be used as oxidising agents, e.g. ozone, peracetic acid. The main requirement is that they are strong oxidisers and that they do not produce toxic end-products. Overall, research shows that ozonation is a very effective tool for elimination of many pharmaceutical residues remaining in treated wastewater (Ternes *et al*., 2003; Andreozzi *et al*., 2004; Huber *et al*., 2005; Baumgarten *et al*., 2007) and in yellow water (Zuleeg, 2005; Tettenbron *et al*., 2007; Gajurel, 2007; Fitzke and Geißen, 2007). Nevertheless, it is very cost-intensive due to the energy required for ozone production and the fact that ozone reacts with all organic compounds present in the liquid and cannot be added explicitly for the pharmaceutical residues. Therefore, as already stated above, a combination of a biological treatment including a lowering of the organic content in the water and an additional ozonation is a promising solution.

The inactivation of pathogens using oxidation by chlorination of the outgoing wastewater is common in many countries. This is generally an efficient method but several factors affect the inactivation of the substances and organisms, mainly other organic substances. Therefore, oxidisation is only a treatment alternative for liquids low in organic material, such as outgoing wastewater and source-separated human urine. A higher content of organic material increases the need for oxidisers, as most are consumed by the organic material present. Furthermore, with high levels of organic material foaming can occur, impairing the treatment process.

## 4. MANAGEMENT OPTIONS.

The risk of 'using' human and animal waste in agriculture needs to be discussed very carefully. First of all, consideration needs to be given to where nutrients and the associated pollutants flows are occurring at the moment. Many believe that the risk to humans is higher from pathogens than from organic pollutants, but both have to be taken into account when estimating the risks of recycling plant nutrients from organic sources.

### 4.1. The status quo of pollutant flows (assuming no degradation).

In the case of solid waste, the pollutant flows depends on the management. If organic waste is collected and treated separately, the nutrients and pollutants are applied in agriculture or horticulture etc. after storage and treatment, e.g. composting. If waste is not separated, the whole waste including its nutrients and pollutants is either dumped, stored in landfills, or burnt in incineration plants.

The main solid fraction of human excreta is deposed of dry, either collected and minimally treated or not collected at all, as 2.6 billion of the world's population do not have access to toilets. Only around one billion people have access to water-based sanitation and of those, only 300 million treat the wastewater to an advanced level. Therefore, the majority of human excreta and wastewater fractions reach recipient waters without any treatment or after some delay on land before stormwater transports them into the recipient waters. When human wastewater is treated, some of the nutrients and pollutants are 'eliminated' by physical, biological and chemical processes in the wastewater treatment plant and the rest are either discharged into rivers and finally into the sea, or captured in the sewage sludge. The sludge is mostly applied to fields or used in soil production, but is sometimes incinerated.

For animal waste, there is hardly any treatment available. Apart from storage, the aim here is to avoid field application in winter. Treatment is only recommended if the animal manure is biologically contaminated or is going to be applied to land in areas where groundwater wells are used for drinking water supply. A pilot plant for aerobic thermophilic manure stabilisation has been tested in Germany (Hahne, 2001) but no further implementation has occurred. The increased interest in bioenergy has increased the interest in farm-based biogas. There, some small amounts of animal manure are anaerobically treated, and in cases of communal digestors within the EU, the animal by-product directives regarding sanitisation of manure (EC 2001/1774 and EC 2006/208) have to be implemented.



The current situation is that potential organic pollutants are applied to arable fields via sewage sludge, compost and animal manure. Another fraction of the pollutants is discharged to water bodies. In wastewater treatment plants, the removal efficiency is dependent on the respective pollutant together with the different technological processes used. For example, Ternes (1998) found removal rates for pharmaceutical residues of between <5-95% in wastewater treatment plants. Hence, the conventional municipal wastewater treatment process is not an efficient barrier for these organic pollutants (Paxéus, 2004; Strenn *et al*., 2004; Castiglioni *et al*., 2006).

Currently, there are only few threshold limits for organic pollutants. In German legislation, PCDF, AOX, PAH, PCBs are monitored in sewage sludge. For compost, animal waste and effluent from communal wastewater treatment plants, there are no thresholds for organic pollutants (BioAbfallverordnung, Klärschlammverordnung).

### 4.2. Option N$^{o.}$ 1: No contamination.

In theory, wastes can be treated in a way that gives zero risk of pollution, for example by incineration or advanced wastewater treatment techniques (e.g. membrane technology followed by UV or oxidation treatment). Both options are high-tech solutions that are very expensive. However, if a zero contamination approach were to be compulsory, then the question is how realistic this option can be. Thousands of incineration plants and high-tech wastewater treatment plants would be necessary to cope with the tremendous amounts of waste and wastewater. For animal excrement this would be a particularly challenging task, as it would require a lot of farm-based treatment plants. Furthermore, in areas with intensive but decentralised animal production, these treatment units would not be suitable.

### 4.3. Option N$^{o.}$ 2: Business as usual.

It is hard to predict whether our current waste management systems will lead to long-term accumulation of organic pollutants in soils and thus compromise soil and water quality. The direct danger for humans is clearly the risk of being infected by pathogens. In developing countries this is a major threat to public health and needs to be tackled urgently. In addition, effects of organic pollutants in water have been noted, e.g. sex changes in fish and increased build-up of antibiotic resistance.

### 4.4. Option N$^{o.}$ 3: Management of pollutant flows.

As mentioned above, the concentrations of nutrients and organic pollutants in waste reflect their sources agriculture, human culture, medicine or industries. Therefore, we need to accept that a certain number of organic pollutants are present in waste outputs from society and that it is impossible to find a zero pollution option. Hence, risk assessments are necessary to identify the inevitable risks emerging from the above-mentioned sources of organic pollutants.

Although there is no final conclusion from research as yet, the following hypotheses have been proposed regarding the potential risks of the organic wastes / excess nutrients:

Organic solid waste: If the waste is composted in a thermophilic process, the hygiene risk is rather low. Organic pollutants with a high volatility are released into the atmosphere and only non-volatile pollutants remain in the compost. Most of the pollutants may be degraded aerobically, but some of them may remain. The potential microbial risk is very low and the potential risk of organic pollutants is rather low as well. The important factor here is mainly process monitoring to ensure that high temperatures are reached and that all of the material is treated.

Human wastewater: In developed countries and in some areas in developing countries, the biological treatment of wastewater is an accepted and established technology. In this case, the organic pollutants are degraded to a certain extent in the wastewater treatment plant and the rest are discharged into recipient water bodies. Some of the organic pollutants are included in the sewage sludge applied to land.

It is strongly recommended that yellow and brown water be source-separated so as to recycle the nutrients without industrial contamination. Without additional treatment, there is a microbial and organic pollutant risk for the separated flows, as the redox status is low and aerobic processes are limited. The microbial risk can be reduced by a variety of measures, the simplest being storage (Vinnerås *et al*., 2008). After treatment the material can be used as a fertiliser in agriculture. If only yellow and black water were applied to fields, then the organic pollutants would be limited to those consumed by humans or produced in the human body (e.g. hormones). It is still an open question whether and how the soil community is affected by these pollutants if they are field-applied on a maximum of three occasions per year. The major concern would be the resistance of microorganisms to antibiotics. If such resistant microorganisms were to find their way back into society, they could massively reduce the efficiency of antibiotic therapy. Their potential accumulation in soil and their uptake by plants also has to be considered. However, there is not enough information available to conclude upon these aspects.

Animal waste: The majority of the animal waste produced is not treated to reduce the risk of microbial or organic pollutants, but the nutrients present in the manure must be used in an environmentally sound way. The potential risks are rather high due to microbial and organic pollutants. Although the amount of antibiotics that are field-applied with liquid manure can be rather large – more than 100 g of tetracycline per hectare and year (Hammer and Clemens, 2007) – there does not seem to be a 'resistance effect', even though the animals may be directly fed on the manured crops.



### 4.5. Management recommendations.

- Consider the treated wastes as an organic or mineral fertiliser. This involves using them to replace mineral fertiliser and avoiding overdoses. This management option limits the flows of organic pollutants. Hammer and Clemens (2007) adapted a model of Rieß (2003) to calculate the flows of organic pollutants depending on the amount of fertiliser applied. The flows from using animal manure were significantly higher than the flows from using source-separated human urine.
- Stop discharging animal waste into water bodies, such as happens in some developing countries, e.g. Vietnam.
- Educate farmers to administer pharmaceuticals according to the recommendations. This avoids high concentrations of their residues in animal manure.
- Educate people that non-used medication should not be disposed via the lavatory. Again, this avoids high concentrations in the waste water.
- Start to source-separate yellow, brown and grey water. This makes it easier to recycle nutrients and to sanitise the different wastewater fractions according to their composition.
- Treat wastewater containing high concentrations of organic pollutants – e.g. in hospitals – with high-tech units.
- Sanitise organic waste, e.g. by thermophilic composting or by ammonia treatment.
- Study the impact of antibiotics on soil communities and water, to test the WHO claim that the soil community may be less affected by organic pollutants than water bodies.

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