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Sewage sludge composting and fate of pharmaceutical residues – recent studies in Estonia

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Abstract. This review is to reflect the work addressed to the application of biosolids and especially sewage sludge as a resource in composting. A considerable drop in the use of P fertilisers can be followed since early 1990s. Due to this fact crop production in Estonia takes place at the expense of soil phosphorous (P) resources. One of the ways of increasing the fertility of agricultural lands is to use nutrient-rich sewage sludge. Unfortunately, this may cause several undesired consequences due to biological and chemical contaminants. The presence of some widely used pharmaceuticals, as ciprofloxacin (CIP), norfloxacin (NOR), ofloxacin (OFL), sulfadimethoxine (SDM) and sulfamethoxazole (SMX), was evident in sewage sludge of the two Estonian largest cities, Tartu and Tallinn. The concentrations of pharmaceuticals decreased after sewage sludge digestion and composting, but they were still present in detectable amounts. Sewage sludge co-composting experiments with sawdust, peat and straw showed the degradation of fluoroquinolones (FQ) and sulfonamides (SA). Additions of sawdust clearly speeded up this process, whereas the mixtures with peat and straw performed lower abilities to decompose pharmaceutical residues. Novel methodologies were developed and experiments conducted to study the potential accumulation of fluoroquinolones FQs and SAs by food plants. Due to the low adsorption of SAs on soil particles they are 'free' to migrate into plants. Different behaviour is characteristic to FQs as they are accumulated in sludge. Recent years have also shown progress in vermicomposting work and in using compost in afforestation.

Key words: composting technologies, fertilizers, pharmaceuticals, plant uptake, sewage sludge, vermicomposting.

INTRODUCTION

Land application of biosolids is generally considered to be the best option of disposal because it offers the possibility of recycling nutrients, provides organic material, improves soil properties, and enhances crop yields (White et al., 2011). Higher soil quality is generally associated with higher concentrations of soil organic matter and a plentiful supply of essential elements. Thus, the recycling of organic matter from anthropogenic residues to soil often benefits agricultural sustainability (White et al., 2013). However, this benefit has to be weighed against potential deleterious effects (White et al., 2011). Whilst recognising its significant value as a resource, recycling sewage sludge to agricultural land requires a careful management to avoid potential negative impacts on the environment from chemical contaminants (Torri et al., 2012).

Organic residues recycling via composting appears to be an ancient activity. The practice of converting animal manure and other biodegradable wastes to compost is believed to have originated as early as agriculture (Fitzpatrick et al., 2005). The earliest known written reference to composting is found in clay tablets dated to the Akkadian empire, about 4,300 years ago (Rodale, 1960), but it is believed that the fertilizer value of aerobically degraded organic matter, which we now call compost, was recognized much earlier. There is evidence that the Romans, Greeks, and the Bani Israel knew about compost. The Bible and Talmud both contain numerous references to the use of rotted manure straw, and mention of compost occurs in 10th and 12th century Arab writings, in medieval Church texts, and in Renaissance literature (Smith et al., 2007).

A worldwide massive use of biosolids as soil conditioners and fertilizers arose in the early 1900s (Frank, 1998). Increasing urbanization and industrialization have resulted in a dramatic growth in the amount of wastes generated globally, particularly of sewage sludge as a byproduct from sewage treatment (White et al., 2011). Land application of treated sewage sludge and other biosolids improves soil fertility and has an important role in closing nutrient cycles (Torri et al., 2012). Among the macronutrients contained in sludge, phosphorus is an essential element for plant metabolism, often considered one of the most limiting nutrients for plant productivity (Shaheen et al., 2012).

A large variety of plant, animal and synthetic wastes can be gainfully composted at scales varying from a household bin to a large industry (Gajalakshmi & Abbasi, 2008). In the composting process, aerobic microorganisms use organic matter as a substrate (Gajalakshmi & Abbasi, 2008). The microorganisms decompose the substrate, breaking it down to more simple compounds (Epstein, 1997; Ipek et al., 2002). During composting, carbon- and nitrogen-containing compounds are transformed through successive activities of different microbes to more stable organic matter, which resembles humic substances (Pare et al., 1998). The rate and extent of these transformations depend on available substrates and the process variables used to control composting (Marche et al., 2003; Gajalakshmi & Abbasi, 2008).

Inventories of soil productive capacity indicate human-induced soil degradation on nearly 40% of the world's arable land (Doran & Zeiss, 2000); this warns us of the ecological collapse of the world's productive soils (Pankhurst et al., 1997). In Estonia the highly industrialised and centralised agricultural production system collapsed in the late 1980s and early 1990s. The area of arable land (crop fields and cultural grasslands) decreased from about one million ha in the early 1990s to less than 0.6 million ha by 2003 (Statistics Estonia, 2006; Iital et al., 2014). Also, a considerable drop in the use of N and P fertilisers took place in the early 1990s when it constituted only about 13% of the peak in 1987–1988. Based on the data from Statistics Estonia in 1994–2001 the average annual consumption of commercial fertilisers was only 85 kg ha^{-1} and in 2009–2011 it reached the level of 120 kg ha^{-1} (Statistics Estonia, 2012; Iital et al., 2014). Since mid–1990s the national average soil P balance has been negative in Estonia due to a sharp decrease in fertilizer use and availability of manure. The national average soil P balance varied in 2004–2009 from -10 to -5 kg P ha^{-1}. Currently crop production in Estonia largely takes place at the expense of soil P resources (Astover & Rossner, 2013).

One of the most efficient ways to eliminate this problem is an intelligent usage of solid waste composts.

This overview is to reflect recent research performed mainly in Estonia in the area of composting. These studies involved different aspects of sewage sludge composting and compost usage; vermicomposting of different waste materials; possible undesired consequences associated with the application of composts in agriculture.

NECESSITY FOR COMPOSTING AND RESOURCES (BACKGROUND)

The soil cover of Estonia is relatively varied due to the alternation of carbonate and humus-rich soils with acid soils which are relatively poor in nutrients and organic matter (Köster & Kõlli, 2013). The lack of nutrients is especially obvious in the case of peatlands which cover 22.3% (10,091 km^2) of Estonia's territory, so restricting the usage of these lands for agricultural purposes. The awareness of the composition and properties of soil cover and its relationship with plant cover in different land use conditions is the basis of ecologically proper and sustainable management of land and soil resource (Köster & Kõlli, 2013).

Estonia has the world's largest exploited oil-shale basin covering about 4% of its territory. In 2001−2013 the number of active landfills in Estonia decreased from 159 to 13. Recultivation of the landscapes covered by semi-coke, oil-shale ash mountains, abandoned opencast mines and closed landfills appears to be one of the major environmental tasks in Estonia.

Biosolids can be used in biofuel production (Raud et al., 2014), leading to the incineration of organic matter. Perceived as a green energy source, the combustion of biosolids has received renewed interest. Still, anaerobic digestion is generally a more effective method than incineration for energy recovery, and digested biosolids are suitable for further beneficial use through land application (Wang et al., 2008). The use of biosolids as a source of organic matter may improve the physical and chemical properties of agricultural soils resulting in an increase in crop yields (Torri et al., 2014). The major potential source for making compost in Estonia is sewage sludge. The yearly generation of sewage sludge by Estonian sewage treatment plants is 30,000 tonnes dw.

Semi-coke is the waste product of oil shale industry and presents the hazard to the environment, due to its phenol and PAH content. One of the main problems of oil shale industry is how to treat semi-coke effectively (Wang et al., 2009). In 1993−2003 the volume of semi-coke formed in Estonian shale oil enterprises varied within 0.6 and 1.4 million tonnes annually (Pae et al., 2005). It has been established that the compost made from semicoke and sewage sludge increases the yield of the crops (Varnik et al., 2006).

The average quantity of biodegradable waste generation in Estonia from grocery stores during 2004–2010 was 9 thousand tonnes year^{-1}. The results of SWOT analysis published by Blonskaja et al. in 2014 showed that composting process is the best solution for kitchen wastes. It has been demonstrated that one of the ecologically and environmentally friendly alternatives to traditional technologies in organic wastes management is vermicomposting, especially in kitchen wastes treatment (Ivask et al., 2013; Peda & Kutti, 2013; Haiba et al., 2014; Sinha et al., 2014).

SEWAGE SLUDGE COMPOSTING AND ENVIRONMENTAL CONCERNS

Unprecedented growth in urban population has resulted in the generation of huge quantities of wastewater worldwide (Singh & Agrawal, 2010). Wastewater treatment facilities are responsible for treating large volumes of domestic and industrial sewage containing human waste. The treatment goal is to produce effluents of high enough quality for discharge back into the environment. Sewage sludge is a byproduct of this process and necessitates proper disposal (Walters et al., 2010; Zuloaga et al., 2012). Safe disposal of sewage sludge is one of the major environmental concerns (Singh & Agrawal, 2010).

Historically, sewage sludge has been disposed of by incineration, landfilling or ocean disposal (Bridle & Skrypski-Mantele, 2000). Nowadays, the most widespread method for sewage sludge disposal has become agricultural application, since it is the most economical outlet for sludge compared to incineration and landfilling (Zuloaga et al., 2012; Li et al., 2013; Chen et al., 2014). The use of sewage sludge in agriculture is one of the major causes of environmental pollution (Nouri et al., 2008). Although, sewage sludge and its compost offers an opportunity to recycle plant nutrients and organic matter to soil for crop production stimulating biological activity (Rodríguez et al., 2012; Zuloaga et al., 2012; Li et al., 2013; Haiba et al., 2014), its usage as a fertilizer is limited due to a large number of toxic pollutants found in this matter (Lillenberg et al., 2010a; Lillenberg, 2011).

Composting is recognized as one of the most important recycling options for sewage sludge (Hara & Mino, 2008; Dorival-García et al., 2015). Since sewage sludge is mainly composted in Estonia and often re-used in agriculture as a fertilizer, several composting methods are applicable, but the selection of the method is dependent on the investment and operation cost, time required to reach compost stability and maturity, the availability of land, origin of raw materials and bulking agents (Ruggieri et al., 2008; Mollazadeh, 2014; Nei et al., 2014).

Several sludge composting experiences have been shared in Estonia (Kanal & Kuldkepp, 1993; Varnik et al., 2006; Kriipsalu et al., 2008; Kriipsalu & Nammari, 2010; Lillenberg et al., 2010a; Holm & Heinsoo, 2013; Kuusik et al., 2014; Menert et al., 2014). The most common sewage sludge composting methods are: static piles, aerated static piles, windrow and in-vessel systems (Yue et al., 2008). There are many factors that affect the composting process, such as the proportions of the mixture, temperature, rate of aeration, oxygen consumption rates, compost pile size, moisture content, pH and carbon-to-nitrogen ratio (Luo et al., 2008; Chen et al., 2014; Malinska et al., 2014; Nayak & Kalamdhad, 2014). Also, microorganisms play a key role in composting processes and nutrient turnover, and even slight changes in microbial activity and community composition due to antimicrobial agents may result in poor compost quality and prolonged time needed for compost stability (Nei et al., 2014). Respiration is a global measure of the total microbial activity that can provide a reliable, repeatable and scientifically sound assessment of microbial activity, respirometry (CO_2 evolution rate and/or O_2 uptake rate) has been widely used to evaluate microbial activity and composting efficiency (Liang et al., 2003; Barrena Gómez et al., 2006). The second widely used parameter for the evaluation of microbial activity is microbial biomass-C, measured by the substrate induced respiration based on Platen & Wirtz, 1999. Also, one of the methods of obtaining information about the dynamics of composting processes is the bacterial-to-fungal ratio (Joergensen & Wichern, 2008). The microbial community may reflect the evolution and performance of the composting process thus acting as an indicator of compost maturity (Nei et al., 2014; Wang et al., 2015).

Since sewage sludge has high moisture content it cannot be composted alone − in order to absorb moisture it should be mixed with dry materials, which act as bulking agent thereby improving the aeration and the compost quality (Nayak & Kalamdhad, 2014; Zhou et al., 2014). Sludge and bulking agent proportions in compost influence the composting reaction rate and the final compost quality. Sludge can be mixed with different bulking agents, sources of carbon, such as peat, straw, wood chips, leaves, ash, peat, sawdust (Komilis et al., 2011; Cukjati et al., 2012; Maulini-Duran et al., 2013; Malinska et al., 2014).

A range of studies has shown that some pharmaceuticals and personal care products (PPCPs) are neither completely removed by sewage treatment, nor completely degraded in the environment (Redshaw et al., 2008; Lillenberg et al., 2009; Lillenberg et al., 2010a; Jelic et al., 2011; Rodríguez-Rodríguez et al., 2012; Borgman & Chefetz, 2013; Haiba et al., 2013b; Narumiya et al., 2013; Reichel et al., 2013). Although, their concentrations are much lower than the levels of traditionally known organic pollutants, the potential long-term effects of these compounds to humans, plants and animals cannot be ignored (Lillenberg et al., 2009; Nei et al., 2014; Van Doorslaer et al., 2014; Prosser & Sibley, 2015; Bártíková et al., 2016).

FATE OF PHARMACEUTICAL RESIDUES DURING SEWAGE SLUDGE COMPOSTING

Pharmaceuticals have been used for decades to prevent and treat human and animal diseases (Zhang et al., 2008; Li et al., 2014). Recently, there has been increasing concern about the effects of pharmaceuticals in aquatic and terrestrial ecosystems, as they can affect the efficiency of microbial-mediated processes (the regeneration of nutrients, carbon and nitrogen circulation and digestion of pollutants) in the environment (Girardi et al., 2011; Jelic et al., 2011; Bergersen et al., 2012; Martín et al., 2012; Chen et al., 2013; Li et al., 2014).

As a result of regular industrial, agricultural and household activities, a variety of compounds enter into the environment, of which only a small percentage are studied for their toxicological effects on humans and the environment (Peysson & Vulliet, 2013). Approximately 4,000 drug substance is used in Europe (human and veterinary), of which may have responsive impact to the environment (Rodríguez-Rodríguez et al., 2011**)**. About 150 medical compounds are studied that have been found in the environment, but mostly in water samples (Rivera-Utrilla et al., 2013; Li et al., 2014). For example, the Estonian Statistics on Medicines data show that over the years the proportion of consumption of different drugs has increased, both over-the-counter as well as prescription drugs (State Agency of Medicines, 2011; 2013). There is no reliable information of how many people actually do or do not consume their drugs, how many medicines are not administered and how many different compounds are thrown into the sewage system or to the garbage. The increasing proportions of administered drugs and personal care products is alarming because of the compound releases to the environment are not controlled (Motoyama et al., 2011; Gonzalez-Martinez et al., 2014), which involves a potential threat to the environment (Vasskog et al., 2009; Rodríguez-Rodríguez et al., 2011; Peysson & Vulliet, 2013).

A wide variety of pharmaceutically active compounds are present in wastewater effluents, surface waters, and ground waters (GWRC, 2008), and the sewage treatment plants are unable to remove all these substances. The removal rates of individual drugs during passage through a sewage treatment plant have varied from 12 to 90% (Stumpf et al., 1999; Butkovskyi et al., 2016). The fate of pharmaceuticals may be divided into three principal routes (Richardson & Bowron, 1985):

1. The substance is ultimately mineralized to carbon dioxide and water;
2. The substance is lipophilic and not readily degradable, so part of the substance will be retained in the sludge. These substances are able to contaminate soil if the sludge is dispersed onto fields;
3. The substance is metabolised to a more hydrophilic form of the parent lipophilic substance, but is still persistent and therefore will pass the sewage treatment plant, ends up in the receiving waters (rivers, seas) and may therefore affect the aquatic organisms, if the metabolites are biologically active.

Presence of different pharmaceuticals in sewage sludge is apparent, but there is still a lack of information concerning the fate of pharmaceutical residues in the environment (Kümmerer, 2008; Lillenberg, 2011). Pharmaceuticals are often not readily degradable (Richardson & Bowron, 1985; Gavalchin & Katz, 1994; Marengo et al., 1997; Halling-Sørensen et al., 2002; Hamscher et al., 2002; Carballa et al., 2004). Still, remarkable amounts of pharmaceuticals enter the soil via fertilizing with sewage sludge (Golet et al., 2002; Haiba et al., 2013a).

Medical substances have many necessary properties to bio-accumulate and provoke change in ecosystems (Kipper et al., 2010; Baran et al., 2011). No trigger values exist for drug residues in sewage sludge neither in Estonia (Decree of Estonian Minister of the Environment) nor in the European Union (EU Council Directive 86/278/EEC; Lillenberg et al., 2009). The most closely related act is the EU directive EMEA/CVMP/055 establishing trigger values for drug residues in manure (EMEA/CVMP/055/96). The content of drug residues should not exceed 100 μg kg^{-1} in manure and 10 μg kg^{-1} in the soil fertilized with manure. Montforts (2005) suggests that these figures should be remarkably lower. Soil organisms, microflora and plants are directly exposed to contaminants in sludge-amended soils.

The presence and content of some widely used pharmaceuticals was determined in sewage sludge and in its compost in the two Estonian largest cities, Tartu and Tallinn (Lillenberg, 2011). The sewage sludge in Tartu was treated by composting – mixing with tree bark (volume ratio 1:1). The methane fermentation and mixing with peat (volume ratio 1:0.75) were used in Tallinn. The samples were taken from anaerobically digested sludge (before mixing with peat) in Tallinn and from untreated sludge (before composting) in Tartu. The concentrations of most of the pharmaceuticals (ciprofloxacin-CIP, norfloxacin-NOR, ofloxacin-OFL, sulfadimethoxine-SDM and sulfamethoxazole-SMX) decreased significantly after sewage sludge digestion and compost processes, but many of them were still present in compost. The degradation of pharmaceutical residues was more efficient in Tallinn probably due to anaerobic sludge digestion (compost was made by mixing the treated sewage sludge with peat) compared to the results obtained in Tartu (raw sewage sludge was mixed with tree bark). The results of the relevant pilot studies are described in detail in Lillenberg et al. (2010a) and Lillenberg (2011).

Interestingly, SDM was present in most sludge and in some compost samples, although this antimicrobial was not marketed any more during the years of 2007 and 2008 in Estonia. It is possible that 'old' supplies were put to use or small amounts of this chemical were imported from other countries (Lillenberg et al., 2010a; Nei et al., 2010).

According to Lillenberg (2011) the highest concentrations of pharmaceuticals were found in Tallinn sewage sludge: CIP 1,520 μg kg^{-1} and NOR 580 μg kg^{-1} (dm). The highest detected concentration of CIP exceeded the trigger value for manure (100 μg kg^{-1}) over four times. The concentrations of OFL (134 μg kg^{-1}), SDM (73 μg kg^{-1}) and SMX (22 μg kg^{-1}) were lower (Table 1). The average contents of antibiotics were: CIP 737 μg kg^{-1}, NOR 279 μg kg^{-1}, OFL 80 μg kg^{-1}, SDM 2 μg kg^{-1} and SMX 18 μg kg^{-1} (dm). As a rule, the concentrations of pharmaceuticals in Tallinn sewage sludge from were relatively low. Still, in some cases the concentrations of CIP, NOR and OFL were over the trigger value (Table 1).

Table 1. The highest concentrations of pharmaceuticals detected from Tallinn sewage sludge, μg kg^{-1} (dm) (reproduced from Lillenberg, 2011)

Month	CIP	NOR	OFL	SDM	SMX
January	**1,520**	**580**	**134**	3	22
February	67	67	17	73	5
March	58	31	8	3	1
April	58	33	3	n.d.	2
May	**150**	**215**	7	0.4	n.d.
June	**206**	**163**	17	n.d.	4
July	39	37	4	n.d.	n.d.
August	11	26	5	n.d.	4
September	0.4	0.4	n.d.	n.d.	n.d.
November	42	16	9	3	3
December	53	85	37	4	7

CIP – ciprofloxacin; NOR – norfloxacin; OFL – ofloxacin; SDM – sulfadimethoxine; SMX – sulfamethoxazole; n.d. – not detected.

In Tartu, contrarily, the concentrations of CIP and NOR were in most cases over the trigger value, the high content of OFL was detected only in August, September and October (Lillenberg, 2011). The content of sulfonamides (SAs – SDM and SMX) was quite low in both cities, under the trigger value set for drug residues in manure (100 μg kg^{-1}) (Tables 1, 2). In Tartu at least one of SAs was present in every sludge sample (Table 2). The contents of SMX were in the range of 0.0–22 μg kg^{-1}, and SDM 0.00–73 μg kg^{-1} (dm) in Tallinn. In Tartu contents of SMX were between 0.0–11 μg kg^{-1}, and SDM 0.0–32 μg kg^{-1} (dm). The highest concentrations of antimicrobials in sewage sludge from Tartu were: NOR – 439 μg kg^{-1} and CIP – 442 μg kg^{-1} (dm). OFL was present in every sludge sample from Tartu and the highest concentration was 157 μg kg^{-1} (dm) (Table 2).

Table 2. The highest concentrations of pharmaceuticals determined from Tartu sewage sludge, $\mu g\ kg^{-1}$ (dm) (reproduced from Lillenberg, 2011)

Month	CIP	NOR	OFL	SDM	SMX
January	**315**	82	86	8	6
February	**423**	**263**	68	32	7
March	89	60	26	0.4	1
May	**174**	**264**	22	1	n.d.
June	**265**	**264**	47	n.d.	16
July	67	**104**	19	n.d.	6
August	**442**	**439**	**111**	24	n.d.
September	**231**	**188**	**157**	22	9
October	**259**	**126**	**149**	4	n.d.
November	**134**	**105**	33	6	11
December	71	40	32	9	6

CIP – ciprofloxacin; NOR – norfloxacin; OFL – ofloxacin; SDM – sulfadimethoxine; SMX – sulfamethoxazole; n.d. – not detected.

The degradation of pharmaceuticals was more efficient in the case of composting in Tallinn. During 12 months composting period the concentrations of all the studied pharmaceuticals diminished for 99.9%, whereas in Tartu this indicator showed the value on average 90 ± 4%. The only exception was SDM, which 'disappeared' fully in both cases. In Tallinn the anaerobically digested sludge was mixed with peat and composted. In Tartu raw sewage sludge was mixed with tree bark (1:1) and settled in piles. The media vas mixed at least twice per month during eight-months period. It has been shown, that a higher decrease of pharmaceuticals is observed after anaerobic digestion than after aerobic digestion, which can be explained by a higher degradation under anaerobic conditions (Martin et al., 2015).

The degradation rate of pharmaceutical residues is dependent on the initial components of the compost. Fine sawdust appears to be an excellent sewage sludge amendment: from the agricultural point of view, sludge co-composted with particularly fine-textured sawdust is claimed to be an excellent compost material to be applied to soils (Ammari et al., 2012; Nei et al., 2015). Kim et al. (2012) have shown that sawdust is able to initiate efficient composting, leading to elevated composting temperatures, and consequently resulting in the reduction of residual concentrations of pharmaceuticals to reasonable levels in a relatively short composting period.

According to Haiba et al. (2013b), composting remarkably reduces the concentrations of these pharmaceuticals. In most experiments their concentrations decreased by 95% or more during 4 months of composting (Table 3). The best results were obtained when the sludge was mixed with sawdust. In the case of using straw or peat instead the decomposition rates were lower. Additions of sawdust clearly speeded up this process, whereas the mixtures with peat and straw performed lower abilities to decompose pharmaceutical residues. No clear evidence was received concerning the impact of oil shale amendments on the degradation speed of the studied pharmaceuticals. Many studies have shown that sawdust has been proven to be a good bulking agent for sewage sludge composting (Banegas et al., 2007; Zorpas & Loizidou, 2008; Haiba et al., 2013a & 2013 b). The decline of tetracycline and sulfonamide concentrations was highly dependent on the presence of sawdust while there was no influence of sawdust on tylosin decline (Kim et al., 2012).

Table 3. Degradation of pharmaceuticals in sewage sludge compost mixtures during 4-months composting period, %

Bulking agent (% from dry matter)	**SMX**	**SDM**	**NOR**	**CIP**	**OFL**
1. peat (50)	83	77	90	92	100
2. sawdust (33)	100	99	96	95	100
3. sawdust + oil shale ash (29+14)	100	96	82	94	99
4. sawdust + wood chips (total 43)	100	99	91	98	86
5. straw (50)	99	98	79	90	74

CIP – ciprofloxacin; NOR – norfloxacin; OFL – ofloxacin; SDM – sulfadimethoxine; SMX – sulfamethoxazole.

PHARMACEUTICALS AND PLANT UPTAKE

The significance of the route involving the uptake of several medicines from soil by plants in terms of risk to human health is evident (Lillenberg et al., 2010b; Prosser & Sibley, 2015; Wu et al., 2015). As the compost made from sewage sludge contains detectable amounts of pharmaceutical residues, experiments were conducted to study the significance of their uptake into plants from soil under 'real' conditions. Therefore, experiments were performed to investigate the potential accumulation of the studied pharmaceuticals – fluoroquinolones (FQs) and sulfonamides (SAs) – taken up by food plants (namely – carrot, potato, lettuce, wheat) from the soil fertilized with sewage sludge or its compost. The results of these experiments are shown in Lillenberg et al. (2010a; 2010b), Kipper et al. (2010) and Nei et al. (2010).

The uptake of pharmaceuticals by the studied food plants was noticeable. It has been shown that due to the low adsorption of SAs on soil particles they readily migrate into plants (Haiba et al., 2013a). Different behaviour is characteristic to FQs due to their sorption to sewage sludge and soil particles (Golet et al., 2003). Therefore, as a rule, the content of SAs in the plants was higher. The content of the studied pharmaceuticals was higher in plats cultivated in sandy soil (Lillenberg, 2011). In loamy soil the molecules of both SAs and FQs attach to clay particles reducing their uptake by plants. Fig. 1 is to illustrate the said. The amounts of FQs going into potato do not depend much on soil type. The application of sewage sludge compost as a fertilizer and the following uptake of pharmaceuticals by food plants may cause contamination of these plants (Haiba et al., 2013a).

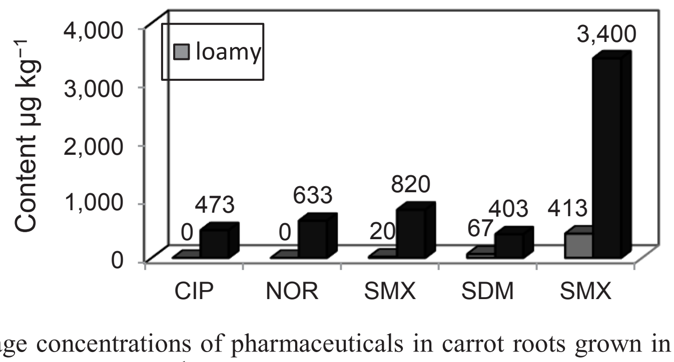

Figure 1. Average concentrations of pharmaceuticals in carrot roots grown in different soils at drug concentration of 10 mg kg^{-1}: CIP – ciprofloxacin; NOR – norfloxacin; OFL – ofloxacin; SDM – sulfadimethoxine; SMX – sulfamethoxazole.

Toxic compounds entering into the soil may affect microbial activity, plant growth and development and may have adverse effects on living organisms (Lillenberg et al., 2010b; Michelini et al., 2012; Haiba et al., 2013a; Nei et al., 2014). Further studies concerning the plant uptake of a wide spectrum of commonly used pharmaceuticals from soils fertilized with sewage sludge or its compost are needed to ensure food safety.

Lillenberg concludes in her PhD thesis (Lillenberg, 2011) that the residues of pharmaceuticals readily accumulate in several food plants. This phenomenon remarkably depends on the nature and concentration of a pharmaceutical and soil type. When using the sewage sludge compost as a fertilizer, it should be carefully tested for the safety. The content of pharmaceuticals in the compost made from sewage sludge may easily lead to the elevated concentrations in food plants if the compost is used as a fertilizer. Still, wheat grains had low or zero concentrations of the analysed pharmaceuticals. This confirmed the potential applicability of sewage sludge compost for fertilization of the crops of this type (Haiba et al., 2013a). Further work should be conducted to determine different types of pharmaceuticals and other organic pollutants by food plants (Lillenberg, 2011). It is evident that the development of novel sewage sludge treatment technologies are needed to solve environmental problems related to sewage sludge exploitation.

PUBLICATIONS AND THESES

Vermicomposting

Vermicomposting technology is a simple and environmentally friendly biological treatment of wastes. As a result of the work published in Ivask et al. (2013) and Haiba et al. (2014) the applicability and efficiency of using earthworms *Eisenia fetida* and *Dendrobaena veneta* in vermicomposting of sewage sludge and household organic residues in the countries with the climate comparable to Estonia was demonstrated.

Compost in afforestation

In Estonia the reforestation of depleted peat and sand mining areas is often complicated due to the unfavourable physical, chemical and biological properties of soils. The impact of artificial roots and soil amelioration with green waste compost in the afforestation of depleted peat fields and sand pits was studied. The results of this work is presented in Jarvis et al. (2012) and Jarvis et al. (2016). Added compost caused significantly improved height growth of the studies tree species seedlings, hence enhanced the growth conditions locally.

Development of novel methodologies for the determination of pharmaceutical residues

Novel approaches for the quantitative determination of traces of commonly used pharmaceuticals in sewage sludge and plants were developed (Lillenberg et al., 2009; Kipper et al., 2011; Kipper, 2012). The compounds were simultaneously extracted from sewage sludge by pressurized liquid extraction (PLE). A novel and effective method for PLE was developed. Solid-phase extraction was used for cleaning up the extracts.

Dissertations defended

PhD thesis: Karin Kipper, Fluoroalcohols as Components of LC-ESI-MS Eluents: Usage and Applications, 2012. A novel and efficient methodology for pharmaceutical analyses in complex matrices (e.g. blood plasma and environmental samples) was developed and tested.

PhD thesis: Merike Lillenberg, Residues of some pharmaceuticals in sewage sludge in Estonia, their stability in the environment and accumulation into food plants via fertilizing, 2011. The aim of the work was to study the presence of some widely used pharmaceuticals in Estonian sewage sludge and its compost and the uptake of these pharmaceuticals from fertilized soils by some food plants. As a result of this research the following was established:

1. Pharmaceuticals were present in sewage sludge and its compost from both Tallinn and Tartu and in several samples their concentrations exceeded the relevant trigger values for manure.
2. Degradation of pharmaceuticals took place as a result of composting.
3. The main reason of the decrease in pharmaceutical concentrations during composting was the applied sludge treatment technology.
4. The uptake of the studied pharmaceuticals by food plants was obvious. The application of sewage sludge compost as a fertilizer and the resulting uptake of pharmaceuticals by food plants may cause contamination of these plants.

CONCLUSIONS

Land application of composts is an important and efficient tool in the remediation of industrial landscapes and agricultural soils in Estonia. Still, due to the frequent presence of different undesired residues, composts made from sewage sludge need careful inspection before their use. The work should be continued by the development of novel and more efficient composting technologies, leading to intelligent solutions of environmental problems related to biowaste exploitation.

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