Sewage Sludge Composting and Pharmaceuticals

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Abstract

Drug residues end up in the environment when sewage sludge or its compost is used as a fertilizer and they cause adverse effects there. Both, the producers and consumers seem to believe that drug residues decompose during sewage sludge treatment or in soil and do not affect the environment or humans. The acceptable level of drugs in different compartments of the environment is still disputable.

Keywords: Pharmaceuticals, Sewage sludge, Compost

Pharmaceuticals in the environment

Intensive use of pharmaceuticals in modern medicine and agriculture is the main reason for global environment pollution by these contaminants (Radović et al., 2016). Widespread occurrence of pharmaceuticals in the environment is well established (Daughton & Ruhoy, 2009). It has now been almost two decades since the defining papers by Halling-Sørensen (1998) and Daughton & Ternes (1999) identified pharmaceuticals in the environment as an important phenomenon. Medical substances have been measured in the effluent of medical care units, sewage and the effluent of sewage treatment plants, in surface water, ground water, and in drinking water (Heberer, 2002). Uncontrolled discharging of these organic compounds into the environmental media has led to their accumulation into soil and sediments (Radović et al., 2016).

Some pharmaceuticals are extremely persistent and introduced to the environment in very high quantities and perhaps have already gained ubiquity worldwide, others could act as if they were persistent, simply because their continual infusion into the aquatic environment serves to sustain perpetual life-cycle exposures for aquatic organisms (Daughton & Ternes, 1999). When drugs are detected in the environment, their concentrations are generally in the ng/L-µg/L (ppt-ppb) range (Moldovan et al., 2009). Even though individual concentrations of any drug might be low, the combined concentrations from drugs sharing a common mechanism of action could be substantial (Daughton & Ternes, 1999). In 2013 Hughes et al. have published a global-scale analysis of the presence of 203 pharmaceuticals across 41 countries and showed that contamination is extensive due to widespread consumption and subsequent disposal to rivers. According to this overview, painkillers were globally the most frequently detected compounds accounting for 31% of records with a median concentration of 230 ng/L followed by antibiotics (21%, 8128 ng/ L).

There are increasing concerns about the undesired impacts that may result from continuous contamination of the environment with pharmaceutically-active substances (Barbosa et al., 2016 and Verlicchi & Zambello, 2016). One of the possible fates of pharmaceuticals is to accumulate in organisms. Bioaccumulation may have different effects from increased internal loads in a given organism potentially reaching toxic concentrations to biomagnification through up-concentration along a food chain (Straub, 2015).

Antibiotics present in soil contaminated with pharmaceutical residues may be taken up by plants from arable land or pasture, and thus involuntarily end up in human or animal food, destroy soil microorganisms or develop drug resistance. Genes determining drug resistance can be transferred from harmless soil microbes to pathogenic microbes (Davies, 1994). It is assumed that using sewage sludge or manure containing drug residues for fertilizing is one of the main reasons of increasing drug resistance (Knapp et al., 2010).

Since 1940 the production and use of antibacterial drugs has multiplied while the antibiotic resistance of bacteria has also increased noticeably. It has been shown that the occurrence of tetracycline resistant gene among soil bacteria increased 15 times between 1970−2008 in the Netherlands, which was caused by using sewage sludge or compost as a fertilizer (Knapp et al., 2010). Probably, the long-term influence of antibiotics on soil microbes has brought about the same effect also in other countries.

Pharmaceuticals in sewage sludge

The environmental presence of pharmaceuticals is attributed primarily to raw or treated sewage (for human drugs) and to manure and lagoons (for veterinary drugs); additional, less obvious sources also exist, which sometimes can play important localized roles (Daughton, 2007). The major route by which pharmaceuticals enter sewage is commonly accepted to be via urine and feces, with each contributing different relative amounts depending on the pharmacokinetics and structure of the individual compound (Winkler et al., 2008). Urban wastewater seems to be the dominant emission pathway for pharmaceuticals globally, although emissions from industrial production, hospitals, agriculture, and aquaculture are important locally

(Beek et al., 2016). Sewage sludge is an inevitable by-product of wastewater treatment. In Estonia about 360,000 − 500,000 tons of it is created annually. Sewage sludge, which is difficult to market, piles up at wastewater treatment plants. Many pollutants are not efficiently removed during sewage and sewage sludge treatment (Martín et al., 2015).

In Lillenberg et al. (2010) the reported highest concentrations (μg/kg) of the antimicrobials norfloxacin (NOR), ciprofloxacin (CIP), ofloxacin (OFL), sulfamethoxazole (SMX) and sulfadimethoxine (SDM) were in sewage sludge as follows: NOR – 162; CIP – 426; OFL – 39; SMX – 6; SDM – 20. In the study carried out by Motoyama et al. (2011), the highest concentrations of the studied pharmaceuticals in sewage sludge were: CIP – 130; SMX – 8; SDM – 3; carbamazepine CBZ – 46. In Martín et al. (2015) the relevant values were: NOR – 258; SMX – 20; OFL – 432; CBZ – 106; and in our recent study (unpublished results): CBZ – 66; diclofenac – 92; triclosan – 1800 (all in μg/kg).

For the prevention of the development of microbial resistance of humans and animals the concentration of antimicrobials in agricultural soil must be clearly under 0.1μg/kg (Lillenberg, 2011). The limited selection of results given above clearly shows that raw sewage sludge in not suitable for improving the quality of agricultural soils.

Fate of pharmaceuticals during sewage sludge composting

Sewage sludge may be regarded as hazardous waste but it can also be used as a fertilizer. Its safety with respect to pharmaceutical residues must be assessed before use (Kipper et al., 2011). Antibiotics are present in Estonian sewage sludge (as elsewhere) and their content may exceed the relevant trigger values for manure (Lillenberg et al., 2011). According to Nayak & Kalamdhad (2015) composting is one of the sustainable practices to convert sewage sludge into useful agricultural product because it is rich in organic matter, micro- and macronutrients, which are essential for plants growth and soil fauna to live. Alternatively, it has been stated that sewage compost cannot be used for agricultural purposes: it may contain an excess amount of chemical contaminants that can be assimilated by food crops (Lillenberg et al., 2010). However, sewage compost is rich in minerals, enabling long-lasting supply for the fast growth of plants (Järvis et al., 2016).

Since the 1960s, Estonia has been the major oil shale producer and consumer in the world (Kalda et al., 2015). 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 (Haiba et al., 2016). 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. 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 these problems is an intelligent preparation of solid waste composts (Haiba et al., 2016).

Sawdust has been proven to be a good bulking agent for sludge composting (Banegas et al., 2007). In the study carried out by Qiu et al. (2012) the degradation of 4 sulfonamides using manure + sawdust or manure + rice straw was more effective than in the case of using manure alone (presumably due to the higher microorganism activities in the former). It has been shown by Kim et al., 2012, that sawdust could be a potential organic source able to initiate efficient composting, as exhibited by elevated composting temperatures, and consequently resulted in the reduction of residual concentrations of tetracyclines, sulfonamides and macrolides to reasonable levels in a relatively short composting period. Thus manure–based composts manufactured through the proper composting process can be acceptable for application to agricultural areas. However, application of livestock manure as raw manure and/or as liquid fertilizer after only a short storage period to stabilize the manure should be avoided as this may result in the potential release of veterinary antibiotics to the environment (Kim et al., 2012).

The degradation rate of pharmaceuticals in sewage sludge compost depends on the applied composting technology. The degradation of salinomycin was observed by Ramaswamyunder et al. (2015) under open and composting conditions. Composting with hay significantly reduced the concentration of salinomycin in the manure, making application of the post-compost manure safer for field application.

It has been shown, that the degradation of fluroquinolones (ciprofloxacin CIP, norfloxacin NOR and ofloxacin OFL) and sulfonamides (sulfadimethoxine SMX and sulfamethoxazole SDM) takes place during sewage sludge co-composting with sawdust, peat and straw (Haiba et al., 2013). Additions of sawdust clearly speeded up the decomposition of the studied pharmaceuticals, whereas the mixtures with peat and straw showed lower abilities to decompose pharmaceutical residues.

In compost mixtures with sawdust the concentrations of the studied pharmaceuticals decreased as much as 95% to 100% during 4-months composting period. The mixtures with straw and peat where less efficient in decomposing these pollutants: in the mixture with peat the degradation level for SMX was 83% and for SDM 76%; in the mixture with straw the degradation level for NOR was 79% and for OFL 74%. At the same time, the concentrations of the other studied pharmaceuticals decreased more than 90% during the 4-month period.

The temperature profiles of the sewage sludge–sawdust mixture samples during composting are demonstrated in figure 1. Initially the temperature of the composting samples ranged from 20 to 38 ^{0}C (mesophilic stage), then rose to 42 ^{0}C in 8 days (start of thermophilic stage).

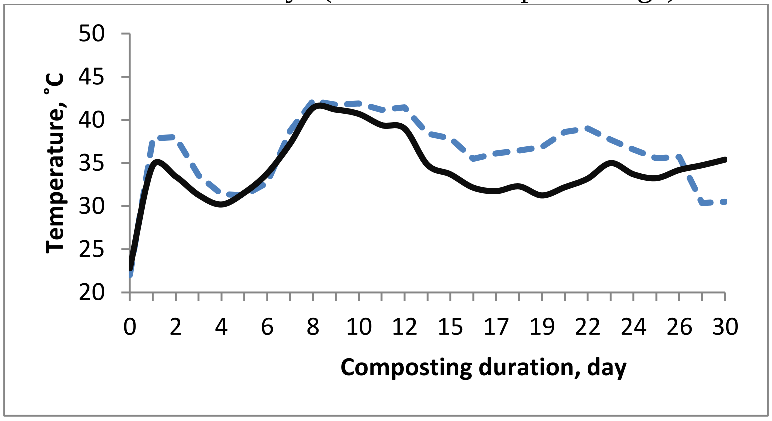

Figure 1. Temperature profiles during sewage sludge composting: sewage sludge mixed with sawdust - - - - E1 1:2 (v:v) and E2 1:3 (v:v)

In small volumes of sawdust-sludge samples, where the sample temperature remained unchanged, the degradation of pharmaceuticals was very slow. During 1-month period only 37% of the initial amount of SMX degraded (the lowest value); the highest level of degradation was apparent in the case of OFL - 82%. This clearly shows that composting may sufficiently speed up the degradation of pharmaceuticals originating from sewage sludge.

Although the results obtained in the case of composting sewage sludge with sawdust seem to be very promising, the experiments show very slow rate of carbamazepine degradation, not exceeding 20% during the 1-month period. According to this fact, the problems associated with the usage of sewage sludge compost as an agricultural fertilizer are far from reaching a final solution.

References:

Astover, A. & Rossner, H. 2013. Knowledge report: Phosphorus status of agricultural soils in Estonia. Baltic Forum for Innovative Technologies for Sustainable Manure Management, p 8.

Banegas, V., Moreno, J.L., Moreno, J.I., García, C., León, G. & Hernández, T. 2007. Composting anaerobic and aerobic sewage sludges using two proportions of sawdust. Waste Management 27, no. 10: 1317–1327.

Barbosa, M.O., Moreira, N.F.F., Riberio, A.R., Pereira, M.F.R. & Silva, A.M.T. 2016. Occurrence and removal of organic micropollutants: an overview of the watch list of EU Decision 2015/495. Water Research 94: 257–279.

Beek, T. aus der, Weber, F.A., Bergmann, A., Hickmann, S., Ebert, I., Hein, A. & Küster, A. 2016. Pharmaceuticals in the environment—Global occurrences and perspectives. Environmental Toxicology and Chemistry 35, no. 4: 823–835.

Daughton, C.G. 2007. Pharmaceuticals in the environment: Sources and their management. In Petrovic, M. & Barcelo, D., eds. Analysis, Fate and Removal of Pharmaceuticals in the Water Cycle, Vol 50. Wilson & Wilson's Comprehensive Analytical Chemistry Series. Elsevier Science, Amsterdam, The Netherlands: 1-58.

Daughton, C.G. & Ruhoy, I.S. 2009. Environmental footprint of pharmaceuticals: The significance of factors beyond direct excretion to sewers. Environmental Toxicology and Chemistry. 28, no. 12: 2495–2521.

Daughton, C.G. & Ternes, T.A. 1999. Pharmaceuticals and personal care products in the environment: agents of subtle change? Environal Health Perspectives 107, suppl. 6: 907–938.

Davies, J. 1994. Inactivation of antibiotics and the dissemination of resistance genes. Science 264, no. 5157: 375–382.

Ferrari, B., Mons, R., Vollat, B., Fraysse, B., Paxēaus, N., Giudice, R.Lo, Rollio, A. & Garric, J. 2004. Environmental risk assessment of six human pharmaceuticals: are the current risk assessment procedures sufficient for the protection of the aquatic environment. Environmental Toxicology and Chemistry 23, no. 5: 1344–1345.

Haiba, E., Nei, L., Ivask, M., Peda, J., Järvis, J., Lillenberg, M., Kipper, K. & Herodes, K. 2016. Sewage sludge composting and fate of pharmaceutical residues – recent studies in Estonia. Agronomy Research 14, no. 5: 1583-1600.

Haiba, E., Nei, L., Lillenberg, M., Kipper, K. & Herodes, K. 2013. Degradation of some pharmaceuticals during composting. AWERProcedia Advances in Applied Sciences 1: 857-862.

Halling-Sorensen, B., Nielsen, S.N., Lanzky, P.F., Ingerslev, F., Lutzhoft, H.C.H. & Jorgensen S.E. 1998. Occurrence, fate and effects of pharmaceutical substances in the environment − a review. Chemosphere 36, no. 2: 357–394.

Heberer, T. 2002. Occurrence, fate, and removal of pharmaceutical residus in the aquatic environment: a review of recent research data. Toxicology Letters 131, no. 1-2: 5–17.

Hughes, S.R., Kay, P., Brown, L.E. 2013. Global synthesis and critical evaluation of pharmaceutical data sets collected from river systems. Environmental Science & Technology 47, no. 2: 661−677.

Järvis, J., Ivask, M., Nei, L., Kuu, A. & Luud, A. 2016. Effect of green waste compost application on afforestation success. Baltic Forestry 22, no. 1: 90-97.

Kalda, K., Ivask, M., Kutti, S., Kuu, A., Meriste, M., Nei, L., Peda, J. & Raukas, A. 2015. Soil invertebrates in semi-coke heaps of Estonian oil shale industry. Oil Shale 32, no. 1: 82–97.

Kim, K.-R., Owens, G., Ok, Y.S.; Park, W.-K., Lee, D.B. & Kwon, S.-I. 2012. Decline in extractable antibiotics in manure-based composts during composting. Waste Management 32, no.1: 110–116.

Kipper, K., Herodes, K., Leito, I. & Nei, L. 2011. Two fluoroalcohols as components of basic buffers for liquid chromatography electrospray ionization mass spectrometric determination of antibiotic residues. Analyst 136, no. 21: 4587-4594.

Knapp, C.W., Dolfing, J., Ehlert, P.A.I. & Graham, D.W. 2010. Evidence of increasing antibiotic resistance gene abundances in archived soils since 1940. – Environmental Science & Technology 44, no. 2: 580–587.

Lillenberg, M. 2011. Residues of some pharmaceuticals in sewage sludge in Estonia, their stability in the environment and accumulation into food plants via fertilizing: PhD thesis in environmental protection. Estonian University of Life Sciences.

Lillenberg, M., Yurchenko, S., Kipper, K., Herodes, K., Pihl, V., Sepp, K., Lõhmus, R. & Nei, L. 2009. Simultaneous determination of fluoroquinolones, sulfonamides and tetracyclines in sewage sludge by pressurized liquid extraction and liquid chromatography electrospray ionization-mass spectrometry. Journal of Chromatography A 1216, no. 32: 5949-5954.

Lillenberg, M., Yurchenko, S., Kipper, K., Herodes, K., Pihl, V., Lõhmus, R., Ivask, M., Kuu, A., Kutti, S., Litvin, S.V. & Nei, L. 2010. Presence of fluoroquinolones and sulfonamides in urban sewage sludge and their degradation as a result of composting. International Journal of Environmental Science and Technology 7, no. 2: 307-312.

Martín, J., Santos, J.L., Aparicio, I. & Alonso, E. 2015. Pharmaceutically active compounds in sludge stabilization treatments: anaerobic and aerobic digestion, wastewater stabilization ponds and composting. Science of the Total Environment 503, special issue: 97–104.

Moldovan, Z., Chira, R. & Alder, A.C. 2009. Environmental exposure of pharmaceuticals and musk fragrances in the Somes River before and after upgrading the municipal wastewater treatment plant Cluj-Napoca, Romania. Environmental Science and Pollution Research 16, suppl. 1: 46-54.

Nayak, A.K. & Kalamdhad, A.S. 2015. Sewage sludge composting in a rotary drum reactor: stability and kinetic analysis. International Journal of Recycling of Organic Waste in Agriculture 4, no. 4: 249-259.

Motoyama, M., Nakagawa, S., Tanoue, R., Sato, Y., Nomiyamac, K. & Shinoharaa, R. 2011. Residues of pharmaceutical products in recycled organic manure produced from sewage sludge and solid waste from livestock and relationship to their fermentation level. Chemosphere 84, no. 4: 432-438.

Qiu, J.R., He, J.H., Liu, Q.Y., Guo, Z.Y., He, D.C., Wu, G.Y. & Xu, Z.C. 2012. Effects of conditioners on sulfonamides degradation during the aerobic composting of animal manures. Procedia Environmental Sciences 16: 17-24.

Radović, T.T., Grujić, S.D., Kovačević, S.R., Laušević, M.D. & Dmkić M.A. 2016. Sorption of selected pharmaceuticals and pesticides on different river sediments. Environmental Science and Pollution Research 23, no. 24: 25232–25244.

Ramaswamy, J., Prasher, S.O., Patel, R.M., Hussain, S.A. & Barrington ,S.F. 2010. The effect of composting on the degradation of a veterinary pharmaceutical. Bioresource Technology 101, no. 7: 2294–2299.

Straub, J. O. 2016. Aquatic environmental risk assessment for human use of the old antibiotic sulfamethoxazole in Europe. Environmental Toxicology and Chemistry 35, no. 4: 767–779.

Verlicchi, P. & Zambello, E. 2016. Predicted and measured concentrations of pharmaceuticals in hospital effluents. Examination of the strengths and weaknesses of the two approaches through the analysis of a case study. Science of the Total Environment 565: 82–94.

Winkler, M., Faika, D., Gulyas, H. & Otterpohl, R. 2008. A comparison of human pharmaceutical concentrations in raw municipal wastewater and yellowwater. Science of the Total Environment 399, no.1-3: 96-104.