

4th International Dry Toilet Conference

Environmental Impact of Micropollutants Present in Urine

*Sanna Pynnönen^{*1} and Tuula Tuhkanen²*

^{*1} corresponding author, M.Sc., Department of Chemistry and Bioengineering, Tampere University of Technology, P.O. Box 541, 33101 Tampere, Finland. E-mail: sanna.pynnonen@tut.fi, Tel: +358 40 198 1144, Fax: +358 3 3115 2869

² Professor of Environmental Engineering, Ph.D., Department of Chemistry and Bioengineering, Tampere University of Technology, P.O. Box 541, 33101 Tampere, Finland. E-mail: tuula.tuhkanen@tut.fi, Tel: +358 40 534 5120

Abstract: Source separated human urine is a valuable and sustainable alternative for mineral fertilizers. Urine contains most of the nutrients coming to wastewater treatment plants and the use of separating toilets can help to reduce this load and to recycle nutrients more efficiently. Yet, the current legislation in the European Union does not directly allow or forbid the use of human urine as a fertilizer. The aim of this paper was to find out more about the possible micropollutants present in urine in developed (Finland as an example) and developing countries. It was discovered that while many pharmaceutical compounds have been detected from the environment in developed countries, practically no data was available on the situation in the developing parts of the world. Source separation of urine and dry toilet technology provide a way to cut the risk chain before possible harmful substances are released untreated into the environment. Dry toilets are also a viable alternative to be used in developing countries.

Keywords: *fertilizer, micropollutant, pharmaceutical, source separation, urine*

Introduction

Every single compound that we humans consume to treat pain, cardiovascular diseases or for example epilepsy, is excreted through our body via faeces or urine. In a modern society where water and wastewater management are well attended, the wastewater produced by a person in a day (approximately 120 litres) is collected into treatment plants and afterwards discharged into receiving waters. This raises one major problem: all the medicines and other pharmaceutical compounds we consume are excreted through our body into urine and faeces, and thus into wastewater. Not everyone has stopped to think about that and the following consequences.

Urine comprises only about 1 % of the wastewater each person produces in a day (Jjemba, 2006), which makes the amount of urine about 1-1.5 litres/person/day. Yet, urine contains most of the macronutrients and micropollutants which arrive as diluted wastewater stream from households to the wastewater treatment plant. Micropollutants are defined as compounds with molecular weight range of 200 to 500/1000 Da (1 Da = 1.66 x 10⁻²⁷ kg) and being present in the aquatic environment at µg or ngL⁻¹ range. (Kümmerer, 2009) Here we refer to pharmaceutical compounds as micropollutants as well.

In the European Union (EU), over 3 000 different substances have been used in medicines (Ternes, 2001). Consequently, many biologically active compounds enter wastewaters and the receiving water bodies without any analyses for specific environmental effects. Municipal wastewater is one of the main exposure routes that bring human pharmaceuticals into the environment: people in private households are either excreting the pharmaceuticals or are improperly disposing of unused or expired drugs into the toilets. To a minor extent, hospital wastewater also contributes to the total loads, though it is highly concentrated and separate treatment for those wastewaters have been studied extensively (Kümmerer, 2001; Kümmerer, 2009). (Zuccato et al., 2000; Ternes et al., 2004)

Urine diversion and diverting toilets (NoMix-technology) allow the use of human urine as an efficient fertilizer. One person produces approximately 2.5-5.7 kg of nitrogen, 0.3-0.6 kg of phosphorous (Mihelcic et al., 2011) and 0.1-1.2 kg of potassium per year (Kirchmann & Pettersson, 1995; Heinonen-Tanski & van Wijk-Sijbesma, 2005). Urine is regarded as sterile unless cross-contamination from faeces occur, which leads to possible persistence of viruses. (Karak & Bhattacharyya, 2011) By utilizing source separated urine the wastewater volume coming to wastewater treatment plants (WWTPs) can be reduced and the nutrient burden arriving to WWTPs can be minimized and redirected: effluent quality is improved, energy savings are gained and nutrients can be recycled better already in their birthplace or near it (Ternes et al., 2004; Ronteltap et al., 2007; Winker et al., 2008; Karak & Bhattacharyya, 2011). Source separation of human urine presents also an opportunity to remove organic micropollutants originating from human metabolism (Maurer et al., 2006).

Yet, the current legislation in the European Union (laws on organic farming, fertilizer products etc.) does not directly allow or forbid the use of human urine as a fertilizer. In Finland, fertilizer regulations don't include the concept urine, only chemically pure urea. (Council Regulation (EC) 2003/2003; 834/2007; 1069/2009) In people's perception, source separated urine might be seen as unsafe or unhygienic compared to cattle manure which contains antibiotics and hormones fed to the animals. And still, only pathogens and heavy metals are considered as harmful substances and therefore analyzed from the manure. Previous studies have tested e.g. the occurrence of pharmaceuticals in Finnish sewage treatment plants and surface waters (Vieno, 2007) and the potential risks of pharmaceuticals in fertilizer use in agriculture (Winker, 2009).

Pharmaceutical compounds are knowingly designed to affect biochemical and physiological functions of biological systems in humans. Nonetheless, they can also elicit biochemical and physiological changes in soil, plants and aquatic organisms. Most of the pharmaceuticals humans consume are excreted via urine (partly via faeces) as unchanged parent compound or as metabolites. (Jjamba, 2006; Jones et al., 2007; Kümmerer, 2009)

Pharmaceutical residues of medicines and hormonal compounds may be excreted through human urine and therefore, usage of urine is associated with risk of transfer of pharmaceutical residues to agricultural fields (Winker et al., 2010). Currently, there are no specific threshold values available for micropollutants in fertilizers (Pronk et al., 2006), but still the introduction of potential hazardous substances into the environment should be avoided. One obstacle for fertilizer use of source separated urine is that there is not enough knowledge about the disadvantageous actions these compounds may elicit in crop plants.

The aim of this paper was to investigate what are the most commonly used pharmaceuticals and hormonal compounds in Finland (per capita per year) that can cause environmental problems directly (improper disposal) or through a secondary exposure via excretion to urine. Also attention was paid on pharmaceutical compounds that are widely used in developing countries and have not yet been properly studied. These micropollutants can be considered as the most important ones to cause endocrine disruptive effects in aquatic animals, induce antibiotic resistance in bacteria or other harmful actions in the environment and thus breaking the risk chain at the premises of the wastewater source is important.

Methods

Pharmaceutical consumption in Finland was studied using statistics gathered by Finnish Medicine Agency, Fimea (2012). The Defined daily dose (DDD) values were obtained from World Health Organization database (WHO, 2012). DDD is specified as the assumed average maintenance dose per day for a drug used for its main indication in adults. By analyzing collected data according to Equation 1

$$C \left(\frac{\text{kg}}{\text{a}} \right) = DDD \text{ (g)} \times \frac{DDD}{1000 \text{ inh}} \times 366 \frac{\text{d}}{\text{a}} \times \frac{\text{inh}}{1000000} \quad (1)$$

the consumed amounts of pharmaceuticals in Finland could be calculated. C is the consumption in kilograms per year, DDD is the defined daily dose, 366 are the number of days in a year and inh is the amount of inhabitants. Also, information on the HIV and tuberculosis situation around the world was examined by searching WHO documents.

Results and Discussion

The selected micropollutants with their consumed amount in Finland in parenthesis included hormonal compounds found in contraceptives (estrogenic hormones, 267 kg a^{-1}) and pharmaceutically active compounds found in epilepsy drugs ($29\ 870 \text{ kg a}^{-1}$), analgesics and anti-rheumatics ($151\ 577 \text{ kg a}^{-1}$), antibiotic quinolones and mycobacterials ($2\ 186 \text{ kg a}^{-1}$), lipid modifying agents ($7\ 266 \text{ kg a}^{-1}$) and beta blockers ($8\ 430 \text{ kg a}^{-1}$), see Figures 1 and 2.

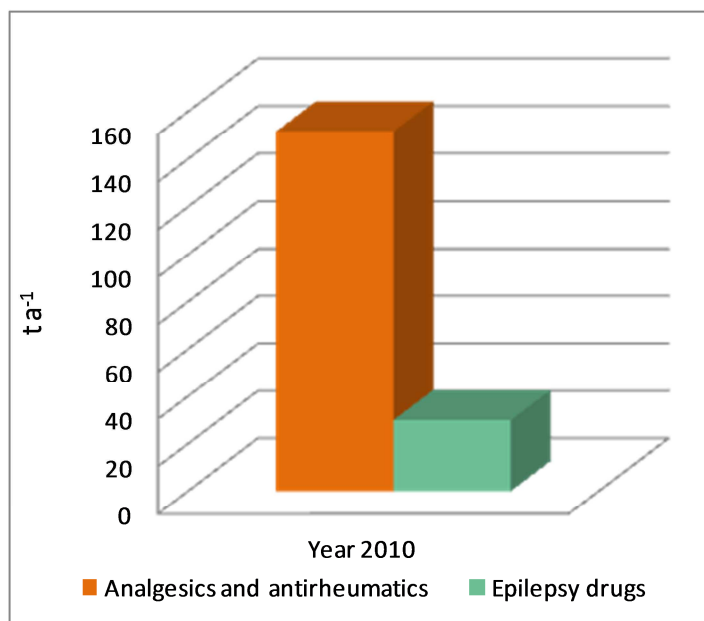


Figure 1. The consumption of both painkillers and antirheumatics, and epilepsy drugs in Finland 2010.

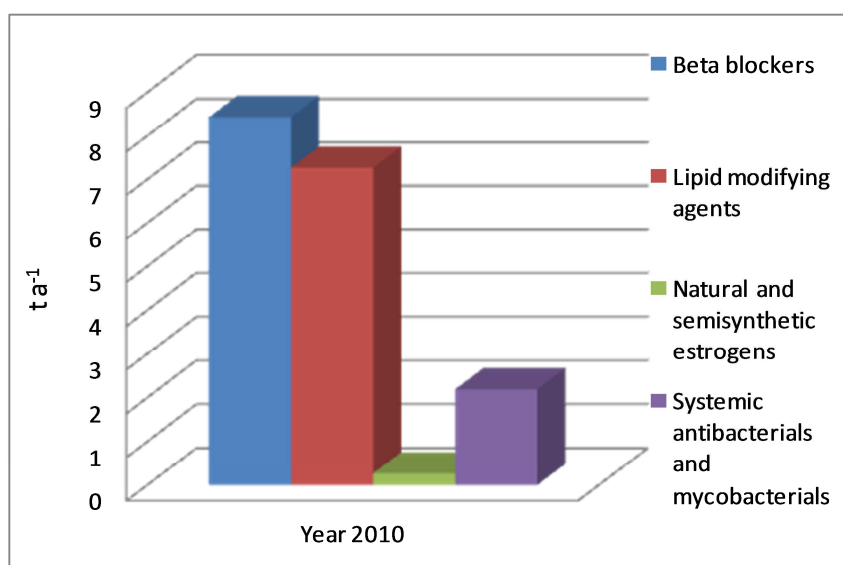


Figure 2. The consumption of beta blockers, anti- and mycobacterials, lipid modifiers and hormones in Finland 2010.

Many of the micropollutants belonging to these categories are excreted to urine as parental compounds or metabolites. In previous studies [e.g. Vieno (2007) and papers related] it has been demonstrated that in developed country such as Finland these compounds can be recovered from environmental matrices, such as surface waters. One of the emerging concerns over the years has been the occurrence of endocrine disrupting compounds (EDCs) such as estrogenic hormones. They are a group of biologically active compounds that are used in both human and veterinary purposes. The adverse health effects of estrogens found in the environment to both animals and humans have been known for some time, and also the harm caused by pharmaceuticals to the ecosystem have been recognized. (Jones *et al.* 2007)

The problem with pharmaceuticals is even bigger in developing countries, where no adequate sanitation system is available. People consume large amounts of antiamebiasis and antiprotozoal pharmaceuticals as well as drugs designed to treat HIV-infections. Yet, practically no information is available on the excreted amounts of these different medical classes and on their effect in the environment.

In the poorer parts of the world HIV-infections (human immunodeficiency virus) are relatively common among the population and pharmaceutical compounds used to treat it include many antiviral and antiretroviral drugs, such as nevirapine, lamivudine and zidovudine. These pharmaceuticals are in most cases used as combinations with other medicines to prevent infections caused by weakened immunology. Such compounds are for instance sulfamethoxazole and trimethoprim, antibiotics commonly used as a cocktail referred as 'cotrimoxazole'. This combination is used by approximately 76 % of all HIV patients in Sub-Saharan Africa (Alcorn, 2010). In addition to those, HIV can lead to emerging of latent tuberculosis which in turn is treated in many cases with combination of antibiotics rifampicin and ciprofloxacin. Tuberculosis infections occur in one third of the HIV-patients (UNAIDS, 2009). The overall infections of malaria, tuberculosis and HIV in the world 2008 are presented in Figure 3.

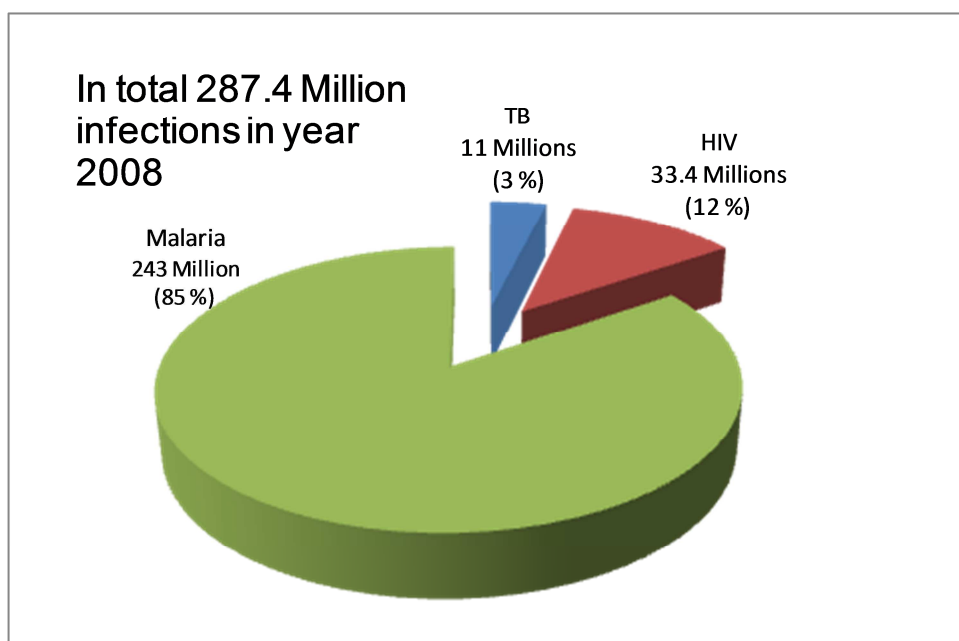


Figure 3. Malaria, tuberculosis and HIV-infections in 2008. (UNAIDS, 2009; Canadian International Development Agency, 2012)

It was discovered from literature that these micropollutants have been detected in aquatic environment on several occasions, the reason being a discharge from wastewater treatment plants. In addition, some studies have reported accumulation of micropollutants in soils that have been irrigated with wastewater. Thus, the micropollutants may migrate into plants and pose risk to edible plants. Trace concentrations of these compounds can cause potential impacts to human health. This is why more knowledge is needed on their negative effects and ways to remove them prior to utilizing source separated urine as a fertilizer. In the case of antiviral drugs, attention has only begun to turn towards them. They have been detected from central European wastewaters and surface waters (Prasse et

al., 2010) and it would appear that they are not removed efficiently in the wastewater treatment plants. Antibiotic resistance of bacteria in wastewater treatment plants has also been a matter of concern for a long time now, but there has been little discussion about the possible resistance of viruses on anti(retro)virals. There are no data available on how these compounds used widely in developing countries will behave in soils and plants or do they inflict negative effects in humans, plants or on other organisms when released into the environment.

Conclusions

It was discovered that while many pharmaceutical compounds have been detected from the environment, practically no data was available on the situation in the developing countries. It is there where the problem with antivirals and antibacterials could be of emerging concern later on. Source separation and dry toilet technology provide a way to cut the risk chain before urine is mixed with clean water and directed to wastewater treatment plants. It also works in countries with less developed infrastructure and this way it might be possible to remove the micropollutants from urine before the environment or humans are exposed. This is yet under study. More knowledge is needed on the (i) negative effects of pharmaceutically active ingredients, the (ii) possible risk caused by leaching and infiltration of them into the groundwater and (iii) ways to remove them prior to utilizing source separated urine as a fertilizer.

References

- Alcorn, K. (2010). TB still killing 4000 people with HIV each day, WHO reports. HIV & AIDS Information. (Accessed 5.4.2012). Available at: <http://www.aidsmap.com/TB-still-killing-4000-people-with-HIV-each-day-WHO-reports/page/1543089/>
- Canadian International Development Agency. (2012). Combat HIV/AIDS, Malaria and Other Diseases (MDG 6). (Accessed 19.3.2012). Available at: <http://www.acdi-cida.gc.ca/acdi-cida/ACDI-CIDA.nsf/eng/JUD-1318912-HFH>
- Council Regulation (EC) 2003/2003. Regulation (EC) No 2003/2003 of the European Parliament and of the Council of 13 October 2003 relating to fertilizers. Available at: <http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2003:304:0001:0194:en:PDF>
- Council Regulation (EC) 834/2007. Regulation (EC) No 843/2007 of the European Parliament and of the Council of 28 June 2007 on organic production and labelling of organic products and repealing Regulation (EEC) No. 2092/91. Available at: <http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2007:189:0001:0023:EN:PDF>
- Council Regulation (EC) 1069/2009. Regulation (EC) No 1069/2009 of the European Parliament and of the Council of 21 October 2009 on laying down health rules as regards animal by-products and derived products not intended for human consumption and repealing Regulation (EC) No 1774/2002 (Animal by-products Regulation). Available at: <http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2009:300:0001:0033:EN:PDF>
- Fimea. (2012). Finnish Medicines Agency. Pharmaceutical consumption data. (Accessed 19.3.2012). Available at (in Finnish): <http://www.fimea.fi/laaketieto/kulutustiedot>
- Heinonen-Tanski, H. and van Wijk-Sijbesma, C. (2005). Human excreta for plant production. *Bioresource Technology* **96**, pp. 403-411

- Jjemba, P.K. (2006). Excretion and ecotoxicity of pharmaceutical and personal care product in the environment. *Ecotoxicology and Environmental Safety* **63**, pp. 113-130
- Jones, O.A.H., Green, P.G., Voulvoulis, N. and Lester, J.N. (2007). Questioning the Excessive Use of Advanced Treatment to Remove Organic Micropollutants from Wastewater. *Environmental Science & Technology* **41**, pp. 5085-5089
- Karak, T. and Bhattacharyya, P. (2011). Human urine as a source of alternative natural fertilizer in agriculture: A flight of fancy or an achievable reality. *Resources, Conservation and Recycling* **55**, pp. 400-4080
- Kirchmann, H. and Pettersson, S. (1995). Human urine - Chemical composition and fertilizer use efficiency. *Fertilizer Research* **40**, pp. 149-154
- Kümmerer, K. (2001). Drugs in the environment: emission of drugs, diagnostic aids and disinfectants into wastewater by hospitals in relation to other sources - a review. *Chemosphere* **45**, pp. 957-969
- Kümmerer, K. (2009). The presence of pharmaceuticals in the environment due to human use - present knowledge and future challenges. *Journal of Environmental Management* **90**, pp. 2354-2366
- Mihelcic, J.R., Fry, L.M. and Shaw, R. (2011). Global potential of phosphorus recovery from human urine and faeces. *Chemosphere* **84** (6), pp. 832-839
- Prasse, C., Schlüsener, M.P., Schulz, R. and Ternes, T.A. (2010). Antiviral Drugs in Wastewater and Surface Waters: A New Pharmaceutical Class of Environmental Relevance? *Environmental Science and Technology* **44**, pp. 1728-1735
- Pronk, W., Palmquist, H., Biebow, M., and Boller, M. (2006). Nanofiltration for the separation of pharmaceuticals from nutrients in source separated urine. *Water Research* **40**, pp. 1405-1412
- Ronteltap, M., Maurer, M. and Gujer, W. (2007). The behavior of pharmaceuticals and heavy metals during struvite precipitation in urine. *Water Research* **41**, pp. 1859-1868
- Ternes, T.A., Joss, A. and Siegrist, H. (2004). Scrutinizing Pharmaceuticals and Personal Care Products in Wastewater Treatment. *Environmental Science & Technology* **38** (20), pp. 392A-399A
- Ternes, T.A. (2001). Analytical methods for the determination of pharmaceuticals in aqueous environmental samples. *Trends in Analytical Chemistry* **20** (8), pp. 419-434
- UNAIDS. (2009). Joint United Nations Programme on HIV/AIDS. Global Facts and Figures. (Accessed 5.4.2012)
Available at: http://data.unaids.org/pub/factsheet/2009/20091124_fs_global_en.pdf
- Vieno, N. (2007). Occurrence of Pharmaceuticals in Finnish Sewage Treatment Plants, Surface Waters, and their Elimination in Drinking Water Treatment Processes. Tampere University of Technology. Publication 666
- WHO. (2012). World Health Organization Collaboration Center for Drug Statistics Methodology. ATC/DDD Index 2012. (Accessed 19.3.2012). Available at: http://www.whocc.no/atc_ddd_index/
- Winker, M. (2009). *Pharmaceutical Residues in Urine and Potential Risks related to Usage as Fertilizer in Agriculture*. Ph.D. Thesis. Hamburger Berichte zur Siedlungswasserwirtschaft, 67. Technische Universität Hamburg-Harburg.
- Winker, M., Clemens, J., Reich, M., Gulyas, H. and Otterpohl, R. (2010). Ryegrass uptake of carbamazepine and ibuprofen applied by urine fertilization. *Science of the Total Environment* **408**, pp. 1902-1908

- Winker, M., Faika, D., Gulyas, H. and Otterpohl, R. (2008). A comparison of human pharmaceutical concentrations in raw municipal wastewater and yellowwater. *Science of the Total Environment* **399**, pp. 96-104
- Zuccato, E., Calamari, D., Natangelo, M. and Fanelli, R. (2000). Presence of therapeutic drugs in the environment. *The Lancet* **355**, pp.1789-1790