





# Pharmaceuticals in municipal sludge – is it real threat or should it be ignored?

Egge Haiba School of Engineering: Tartu College

08.02.2018

## **RESEARCH TEAM**

		Lembit Nei	<ul> <li>Professor, Director of Tartu College - Chemistry and Chemical Technology; SPECIALITY: Environmental Chemistry</li> <li>Tallinn University of Technology, School of Engineering, Tartu College, Tartu College of TUT</li> </ul>
		Merike Lillenberg	<ul> <li>Associate Professor - FIELD OF RESEARCH: Biosciences and Environment; Food Sciences; Research relating to the State of the Environment and to Environmental Protection</li> <li>Estonian University of Life Sciences, Institute of Veterinary Medicine and Animal Sciences</li> </ul>
		Koit Herodes	<ul> <li>Associate Professor in Analytical Chemistry, Head of Institute of Chemistry - Chemistry and Chemical Technology; SPECIALITY: Chromatographic and mass-spectrometric methods of analysis (LC-MS and GC-MS); Solvents effects, their influence on physico-chemical properties and reactions</li> <li>University of Tartu, Faculty of Science and Technology, Institute of Chemistry</li> </ul>
		Karin Kipper	<ul> <li>PhD, Research Fellow in Analytical Chemistry -</li> <li>University of Tartu, Faculty of Science and Technology, Institute of Chemistry.</li> </ul>
of Technolo		Mari Ivask	<ul> <li>Professor -Ecology, Biosystematics and –physiology; Agricultural Sciences; Research relating to the State of the Environment and to Environmental Protection.</li> <li>Tallinn University of Technology, School of Engineering, Tartu College</li> </ul>
		Egge Haiba	<ul> <li>PhD - Biosciences and Environment; 1.9. Research into Substances Hazardous to the Environment; SPECIALTY: Environmental technology, pollution control.</li> <li>Tallinn University of Technology, School of Engineering, Tartu College</li> </ul>
		Jane Raamets	<ul> <li>MSc, PhD student -Research relating to the State of the Environment and to Environmental Protection; SPECIALTY: Environmental technology, pollution control, soil mechanics.</li> <li>Tallinn University of Technology, School of Engineering, Tartu College</li> </ul>
		Sander Kutti	<ul> <li>MSc, PhD student - FIELD OF RESEARCH: Biosciences and Environment; Research relating to the State of the Environment and to Environmental Protection</li> <li>Tallinn University of Technology, School of Engineering, Tartu College</li> </ul>

### **Composting studies**

The presence and degradation of pharmaceuticals -

- 1. in sewage sludge and its compost;
- 2. plant uptake of selected pharmaceuticals from fertilized soils;
- 3. the bulking agent effect on the degradation of pharmaceutical residues present in sewage sludge compost;
- 4. the impact of these pharmaceuticals on microbial activity.





## Pharmaceuticals

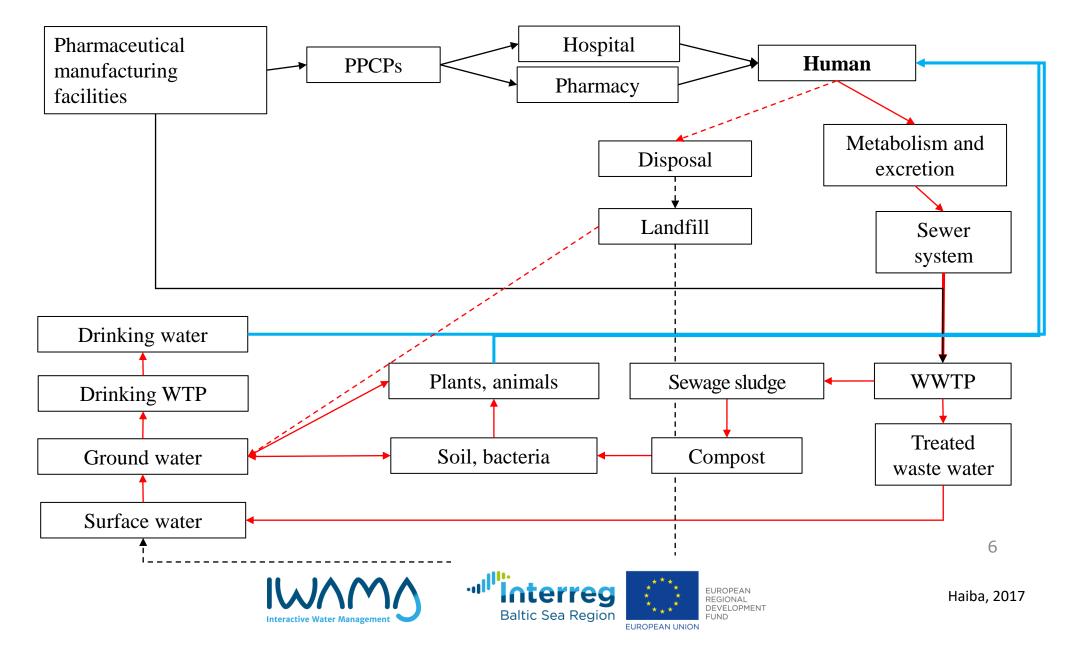
- Purpose prevention and treatment of diseases
- Effect on humans/animals weak knowledge
- Degradation /no degradation + metabolites



- ~4,000 drug substance is used in Europe (human and veterinary),
- ~250 medical compounds are studied (mostly found in the water environment),
- consumption -> no reliable information,
- releases to the environment are not controlled -> a potential threat to the environment
- the most frequently occurring drug concentrations between
  - ng/L to low  $\mu\text{g/L}$  in treated wastewater
  - $\mu$ g/kg to low mg/kg (dry weight) in biosolids



#### Fate of human pharmaceuticals in the environment





# Pharmaceuticals in sewage sludge

Contaminants (Treated sewage sludge):

- Heavy metals
- Fecal coliforms
- Helminths eggs

Trigger value



EU Council Directive

86/278/EEC, 1986

Other:

- PHARMACEUTICALS ----- NO trigger value
- EMEA/CVMP (1996) the content of veterinary medicines should not exceed 100  $\mu g~kg^{-1}$  in manure, and 10  $\mu g~kg^{-1}$  in soil fertilized with manure
- Recommended value considerably lower 1  $\mu g~kg^{\text{-1}}$  for the soil and 0,4 ng L^1 for the water compartment





## Pharmaceuticals in sewage sludge



- Pharmaceuticals are present in different concentrations in effluents and biosolids.
- Some pharmaceuticals do not decompose during sewage sludge treatment
- Sewage treatment facilities do not remove all pharmaceutical residues completely.
- Effluent and biosolids are reused in agriculture for irrigation and fertilisation.
- In soil APIs can affect microorganisms, accumulate in plants and may have adverse effects on living organisms.



## Sewage sludge as fertilizer

A major public concern regarding agricultural applications of treated wastewater and biosolids is the introduction of contaminants such as PPCPs from these "waste streams" to crops via plant uptake.









#### Pharmaceuticals in plants (Lillenberg, 2011)



Lettuce samples.



Potato samples



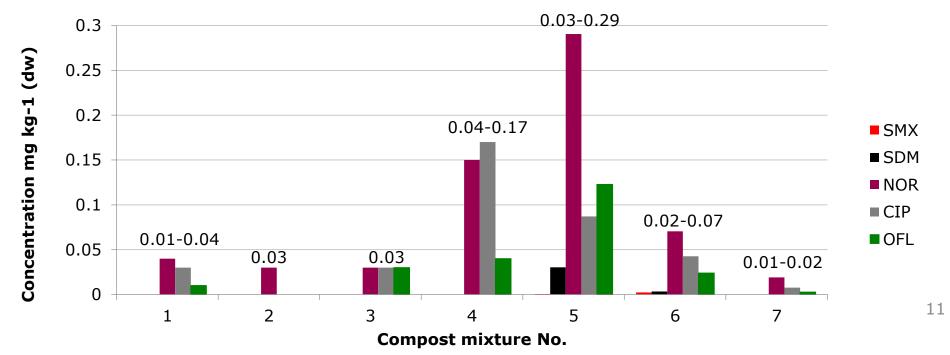


## Initial content of antibiotics in sludge (Haiba, 2017)

• Antibiotics tested:

SMX - sulfamethoxazole, SDM - sulfadimethoxine, NOR - norfloxacin, CIP - ciprofloxacin and OFL - ofloxacin

• Contents of pharmaceuticals were detected in fresh compost mixtures.





## Pharmaceuticals in sludge and compost (Haiba, 2017)

• PPCPs tested:

DFC - diclofenac, CBZ - carbamazepine, MET – metformin and TCS - triclosan.

PPCP	Mixture ratio (v:v)	Before spiking	1 day	1 week	1 month
	1:2	$0.09 \pm 0.00$	2.65±0.32	1.31±0.04	0.21±0.01
DCF	1:3	$0.06 \pm 0.00$	2.38±0.21	$1.37 \pm 0.04$	$0.04 \pm 0.00$
СВΖ	1:2	$0.06 \pm 0.00$	3.11±0.38	$2.59 \pm 0.05$	$3.20 \pm 0.01$
	1:3	$0.05 \pm 0.00$	2.69±0.26	2.31±0.08	2.32±0.08
МЕТ	1:2	$0.00 \pm 0.00$	2.14±0.25	0.44±0.02	$0.18 \pm 0.01$
	1:3	$0.00 \pm 0.00$	$1.95 \pm 0.15$	0.23±0.02	$0.14 \pm 0.02$
TCS	1:2	1.77±0.06	4.54±0.38	3.24±0.20	2.07±0.12
	1:3	1.23±0.07	3.53±0.14	2.54±0.09	$0.68 \pm 0.02$



## On the degradation of some pharmaceuticals (Haiba, 2017)

PPCP	Mixture ratio (v:v)	Haiba, 2017			Data from literature		
		k ( d-1)	t <sub>1/2</sub> (d)	%	k ( d-1)	t <sub>1/2</sub> (d)	%
МЕТ	1:2	0.22	3	91	0.12 0.26 <sup>A</sup>	1 5 <sup>A</sup> 2 3 <sup>C</sup>	99100 <sup>A</sup>
	1:3	0.27	3	93	0.22 0.27 <sup>c</sup>		
	1:2	0.09	7	92	0.23 0.16 <sup>A</sup> 0.010 <sup>B</sup>	3 4 <sup>A</sup> 70 <sup>B</sup>	26 <sup>в</sup>
DCF	1:3	0.09	8	98			
тсѕ	1:2	0.03	26	55	0.05 0.04 <sup>A</sup>	13 20 <sup>A</sup> 35 <sup>B</sup>	
	1:3	0.05	13	81	0.02 <sup>B</sup>		45 <sup>B</sup>
CBZ	1:2	0.00	222	-11		46173ª	
	1:3	0.00	178	13			

<sup>A</sup> agricultural soil

<sup>B</sup> sterile soil

<sup>C</sup> compost mixture





13

# Pharmaceuticals in the environmentwhat is the threat?

- Some pharmaceuticals degrade, some are persistent
- Highly persistent APIs are able to accumulate in plants and animals.
- High levels of APIs in the environment dangerous to water and soil organisms.
- Low drug levels in the environment soil and water microbes develop drug resistance.
- High drug levels in food (animal feed) destructive to plants, dangerous to human (animal) health.
- Low drug levels in food (animal feed) -

the development of drug-resistant microbes in humans or animals.







# **Clear waters from pharmaceuticals - CWPharma**

errec

EUROPEAN UNION

**EVELOPMENT** 

IROPFAN

Baltic Sea Region

- works on reducing information gaps regarding :
  - sources,
  - emissions and
  - environmental concentrations of APIs in the Baltic Sea.
- The project evaluates different API emission reduction measures.
- The best existing practices of the partner countries shall be shared in order to promote the sustainable management of APIs in the Baltic Sea region.
- <u>http://www.syke.fi/en-</u> <u>US/Research</u> <u>Development/Research</u> <u>and</u> <u>development</u> <u>projects/Projects/Clear</u> <u>waters</u> <u>fr</u> <u>om</u> <u>pharmaceuticals</u> <u>CWPharma</u>





# Thank you for your attention!

TALLINN UNIVERSITY OF TECHNOLOGY School of Engineering: Tartu College Puiestee 78, 59001 Tartu, Estonia egge.haiba@ttu.ee