For our Environment

17th IWA International Conference on Diffuse Pollution and Eutrophication

Antibiotics in Groundwater Under Locations with High Livestock Density in Germany

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1. Background

Antibiotics:

• Type of antimicrobial that may either kill or inhibit the growth of other bacteria.

• Most important for treatment and prevention of bacterial infections.

• In 2014 1.238 tons of antibiotics were supplied to veterinarians (650 tons of antibiotics in human medicine)

• Antibiotics, their metabolites and transformation products as well as antibiotic-resistant bacteria (MRSA, ESBL) were detected in the environment worldwide.

• The high consumption of antibiotics by men and animals and the resulting effects for man and environment are an issue of increasing global concern.
2. Pathways of pharmaceuticals into the environment

- Optimized to stability and mobility, and not easily bio-degradable
- Up to 80% are excreted and enter the environment via different pathways
- Human pharmaceuticals enter the municipal sewer system and end up in sewage treatment plants.
- With treated wastewater they enter surface waters or via the disposal of sewage sludge on agricultural soils
- Veterinary pharmaceuticals release into the environment by spreading manure
- After percolation through the soil they may reach shallow groundwater, adjacent surface water bodies via runoff from fertilized farmlands.

<table>
<thead>
<tr>
<th>Substance</th>
<th>Concentration in Groundwater</th>
<th>Publication</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sulfadiazine</td>
<td>&lt; 0,1 - 0,16 µg/l</td>
<td>Alexy, Kümmerer (2005)</td>
</tr>
<tr>
<td></td>
<td>0,01 µg/l</td>
<td>UBA (2014)</td>
</tr>
<tr>
<td>Sulfadimidine</td>
<td>max. &gt; 0,2 µg/l</td>
<td>Höper et al. (2011)</td>
</tr>
<tr>
<td></td>
<td>max. 0,16 µg/l</td>
<td>Hirsch et al. (1999)</td>
</tr>
<tr>
<td></td>
<td>max. 0,21 µ/l</td>
<td>BLAC (2003)</td>
</tr>
<tr>
<td></td>
<td>max. 0,24 µg/l</td>
<td>Hamscher et al. (2004)</td>
</tr>
<tr>
<td></td>
<td>max. 4,00 µg/l</td>
<td>Weiss (2008)</td>
</tr>
<tr>
<td></td>
<td>0,011 µg/l</td>
<td>UBA (2014)</td>
</tr>
<tr>
<td>Sulfamethoxazole</td>
<td>max. 0,3 µg/l</td>
<td>LANUV NRW (2008)</td>
</tr>
<tr>
<td></td>
<td>max. 0,95 µg/l</td>
<td>UBA (2014)</td>
</tr>
<tr>
<td>Tetracycline</td>
<td>max. 0,13 µg/l</td>
<td>Hamscher et al. (2005)</td>
</tr>
<tr>
<td>Tylosin</td>
<td>1,0 µg/l</td>
<td>Weiss (2008)</td>
</tr>
<tr>
<td>Trimethoprimeth</td>
<td>0,04 µg/l</td>
<td>LfW (2004)</td>
</tr>
<tr>
<td>Amoxicillin</td>
<td>0,1 µg/l</td>
<td>BLAC (2003)</td>
</tr>
</tbody>
</table>
3. Goal of the project

- The extent of groundwater pollution by antibiotic residues from livestock farming is unknown in Germany.

- Monitoring programs or a threshold value for pharmaceuticals in groundwater do not exist.

- The Federal Environment Agency supported a project that aimed to investigate near-surface groundwater samples in regions of high livestock density (stocking rates) and a high risk of groundwater exposure to antibiotics.

- Therefore worst case criteria were developed in order to identify suitable sampling sites within the existing groundwater monitoring grid and to foster a high probability of leaching of antibiotics to the groundwater.
4. Material and Methods
4.1 Selection of veterinary antibiotics

Application in study area

Substance-specific Characteristics

Findings in Literature

Results

23 Einzelwirkstoffe
- Sulfaethoxypyridazine
- 4-OH-Sulfadiazine
- Sulfadiazine
- Sulfathiazole
- Trimethoprim
- Sulfadimidine
- Sulfadoxine
- Sulfamethoxypyridazine
- N-Ac-Sulfadiazine
- Sulfachloropyridazine
- Sulfamethoxazole
- Sulfadimethoxine
- Tetrazykline
- 4-epi-Tetrazykline
- Oxytetracykline
- 4-epi-Oxytetracykline
- Chlortetrazykline
- 4-epi-Chlortetrazykline
- Enrofloxacine
- Lincomycin
- Tilmicosin
- Tulathromycin
- Erythromycin

• Carbamazepine was chosen as a tracer for inputs of human medication
4.2 Selection of study area and sampling points

Selection criteria:
- Very high livestock density and intensive application of manure and slurry
- Sandy soils with low gross field moisture capacity
- Low natural protective function of the near-surface groundwater layer
- Low depth of groundwater surface (< 5 meters below surface)
- Very high nitrogen concentrations in groundwater (nitrate and/or ammonium) above EU thresholds
4.3 Sample collection, preparation and analysis

• At least two groundwater samples were taken from each sampling point in 2012 and 2013.

• **Groundwater samples** were stored in glass bottles and delivered to the laboratory immediately.

• Groundwater samples were analyzed with multi-methods, based on solid phase extraction as well as liquid chromatographic separation coupled to detection by mass spectrometry (LC-MS-MS).

• The limits of detection respectively quantification were on a concentration level of 1-15 ng/l for all 23 single substances.

• Besides antibiotics also the main and accessory solutes were analyzed in order to characterize the groundwater.
5. Results and Discussion

With regard to the 48 investigated groundwater measuring points:
• At 39 locations no detections of veterinary antibiotics were ascertained;
• At 7 locations sulfonamides were detected at low concentrations <100 ng/L;
• At 2 locations sulfonamides were detected at high concentrations >>100 ng/L.

Substances
- Sulfaethoxypyridazin
- 4-OH-Sulfadiazin
- Sulfadiazine
- Sulfathiazole
- Trimethoprim
- Sulfadimidine
- Sulfadiazine
- Sulfamethoxazole
- Sulfadoxin
- N-Ac-Sulfadiazin
- Sulfachloropyridazin
- Sulfamethoxypyridazin
- N-Ac-Sulfadiazin
- Sulfadimethoxin
- Tetrazyklin
- 4-epi-Tetrazyklin
- Oxytetrazyklin
- 4-epi-Oxytetrazyklin
- Chlortetrazyklin
- 4-epi-Chlortetrazyklin
- Enrofloxacin
- Lincomycin
- Tilmicosin
- Tulathromycin
- Erythromycin

Sampling Sites
Results
- < Limit of Detection (LOD)
- > Limit of Detection
- > Limit of Quantification (LOQ)
- > 0.1 μg/L

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6. Conclusions

• Based on the worst-case approach we concluded that the leaking of veterinary antibiotics into shallow groundwater is not ubiquitous in Germany.

• Unfavourable conditions of single locations show a partly high emission of antibiotics with a high seasonal variability.

• Specific sources of deposition could not be determined and discovered during this project.

• Research is presently being extended and focused on the clarification of sources with intensive samplings in groundwater and also slurry.

• As a matter of precaution we recommend a threshold value of 0.1 micrograms per litre (µg/l) for pharmaceuticals in groundwater and also monitoring programs. In case of exceedence there would be a legal basis to introduce adequate measures to protect groundwater.
7. Further Information


Thank you for your attention

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