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Managed aquifer recharge: the fate of pharmaceuticals from infiltrated treated wastewater investigated through soil column experiments

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The EU FP7 project MARSOL addresses water scarcity challenges in arid regions, where managed aquifer recharge (MAR) is an upcoming technology to recharge depleted aquifers using alternative water sources. Within this framework, column experiments were conducted to investigate the fate of pharmaceuticals when secondary treated wastewater (TWW) is infiltrated through a natural soil (organic matter content 6.8%) being considered for MAR. Three parallel experiments were run under conditions of continuous infiltration (one column) and wetting-drying cycles (two columns, with different analytes) over a 16 month time period.

The pharmaceuticals diclofenac, ibuprofen, carbamazepine, naproxen, gemfibrozil, and fenoprofen, as well as the antibiotics doxycycline, sulfadimidine, and sulfamethoxazole, are commonly present in treated wastewater in varying concentrations. For the experiments, concentration variability was reduced by spiking the column inflow water with these compounds. Concentrations were periodically analyzed at different depths in each column and the mass passing each depth over the duration of the experiment was calculated.

At the end of the experiments, sorbed pharmaceuticals were extracted from soil samples collected from different depths. A pressurized liquid extraction method was developed and resulted in recoveries from spiked post-experiment soil samples ranging from 64% (gemfibrozil) to 82% (carbamazepine) for the six non-antibiotic compounds. Scaling results by these recovery rates, the total mass of pharmaceuticals sorbed to the soil in the columns was calculated and compared to the calculated attenuated mass (i.e. mass that left the water phase). The difference between the attenuated mass and the sorbed mass is considered to be mass that degraded.

Results for continuous infiltration conditions indicate that for carbamazepine and diclofenac, sorption is the primary attenuation mechanism, with missing (i.e. degraded) mass lying within the propagated measurement error range. Over the duration of the experiment, 36% of carbamazepine and 59% of diclofenac passed the deeper sediment (depth 71 cm, last sampling point along an 88 cm flowpath through soil) in the water phase. On the other hand, the compounds fenoprofen, gemfibrozil, ibuprofen and naproxen showed degradation rates (degraded relative to total infiltrated mass) of 51%, 57%, 63% and 95%, respectively. Corresponding results for wetting-drying cycles (one column with antibiotics spiked and analyzed, one without) will also be presented, where oxygenated conditions during drying periods and may influence degradation.

The results indicate that while substantial portions of mass degrade for some compounds, sorption is also an important mechanism for mass leaving the water phase. Although the most sorbed mass is present near the surface, substantial amounts of mass also sorb at depth. A flowpath through a thick unsaturated zone composed of a soil favorable to sorbing polar organic compounds presents the best chance to attenuate the most mass, but consequently micropollutants will accumulate in the soil if degradation remains low and re-mobilization of the compounds may occur when system conditions change. However, the results of these experiments also suggest that for the chosen soil and infiltration conditions, near-complete degradation of fenoprofen, gemfibrozil, ibuprofen and naproxen is possible considering a substantial unsaturated zone thickness.