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Redox-sensitivity and mobility of selected pharmaceutical compounds in a laboratory column experiment

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Laboratory column experiments are suitable to investigate the sediment water interaction and to study the transport behaviour of solutes. Processes like retardation and degradation can be identified and quantified. The conducted experiment, which is closely connected to a field study in Luxembourg, investigated the transport behaviour of selected pharmaceutical compounds and their redox-dependent metabolism under water saturated conditions.
Fine-grained natural sediment with a low hydraulic conductivity from a study site in Luxembourg was filled into the column. The water for the experiment was taken from a small stream at the same fieldsite. It was spiked with four pharmaceutical compounds (carbamazepine, diclofenac, ibuprofen, sulfamethoxazole) with concentrations between 170 and 300 ng/L for the different substances. The chosen pharmaceuticals were also detected in groundwater and surface water samples at the study site and used to qualify exchange/mixing of surface water and groundwater (BANZHAF et al., 2011). As some of the substances are known to exhibit redox-sensitive degradation, the redox-conditions were systematically varied throughout the experiment. This was realised by adding nitrate at the inflow of the column. During the experiment, which lasted for 2.5 months, four different nitrate concentrations (20-130 mg/L) were applied, beginning with the highest concentration. During the experiment water from the reservoir tank was sampled daily in order to detect a potential degradation of the pharmaceutical compounds before they enter the column. The effluent water was sampled every three hours to guarantee a maximum resolution for the analysis of the pharmaceuticals where necessary. In addition, major ions were analysed in the influent and effluent samples. Throughout the experiment physicochemical parameters (oxidation reduction potential (ORP), dissolved oxygen, electrical conductivity, and pH-value) were measured and logged at the outflow of the column.
At the beginning, the ORP was positive (200 mV) and then dropped continuously. Negative values were reached after 1 month and at the end of the experiment -300 mV were measured. Apart from nitrate and nitrite no significant changes in ion concentrations were detected in the effluent. However, the added pharmaceuticals showed very different behaviour in the column. Diclofenac and especially carbamazepine were highly absorbed by the sediment. They were detected significantly later at the outflow of the column than sulfamethoxazole and ibuprofen.
Sulfamethoxazole was heavily influenced by the redox-conditions. Its time variation curve in the effluent is negatively correlated with nitrite and nitrate: during nitrite formation the concentrations of sulfamethoxazole dropped considerably.
The presented experiment yields a better understanding of the processes influencing the occurrence and transport behaviour of the studied compounds. In addition, some general findings on redox-dependent transport behaviour and metabolism of the antibiotic sulfamethoxazole are gained. This emphasizes the role of the ORP as a key parameter for the behaviour of this compound, which has to be considered.

BANZHAF, S., KREIN, A. & SCHEYTT, T. (2011). Investigative approaches to determine exchange processes in the hyporheic zone of a low permeability riverbank. Hydrogeology Journal 19 (3), pp. 591-601.