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Investigation of pharmaceutical transport in saturated sandy aquifers using column experiments: the effect of pH

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Due to the development of advanced analytical techniques it is increasingly known that a high number of polar organic trace compounds, particularly residues of pharmaceuticals, occur in the aquatic environment. In contrast to the sources and pathways of such compounds, their impact on ecosystems and their fate in different environmental compartments are comparatively less investigated. Because of the spatial extension and time available, the zone between water and natural solids (e.g. sediments or soil in groundwater zones, bank filtration sites and for soil aquifer treatment) plays an important role in the elimination of anthropogenic trace compounds from water phase. Here, degradation and sorption processes mainly influence the content of trace compounds.
Correlations, specific for compound groups, between n-octanol-water distribution coefficients, available from experiment or calculations, and sorption coefficients (e.g. KOC) often allow a suitable prognosis of the transport behavior of organic pollutants in an underground passage. In case of polar, ionizable organic compounds such prediction is problematic and often not possible. Here, besides relatively weak non-polar van der Waals attraction, other interaction mechanisms, such as covalent bonding, complex formation, or ion exchange, can dominate. The latter is closely connected with the type of basic and/or acid groups in a molecule. The degree of protonation could be changed in dependence of type and concentration of other ions and of the acidity constants (pKa) and therefore from pH.
Laboratory column studies at different pH value (range from 4 to 8) were carried out using natural sandy sediments from aquifers and model water containing selected pharmaceuticals to investigate the influence of degree of protonation on sorption. Eight different pharmaceuticals were chosen for laboratory column experiments. Their selection was based on the presence of basic/acid functional groups, pKa, high production and consumption rates, and occurrence in environment. The long-term objective of this research is to consider specific interactions such as ion exchange for the improved transport models.
Breakthrough experiments show that retardation is significantly influenced by pH for the majority of the selected pharmaceuticals. As a general tendency, it was observed that a decreasing pH caused an enhanced delay. For acidic compounds such as naproxen, this behavior was expected because of the neutral species being the dominating one. The stronger retardation of cationic agents such as atenolol with decreased pH could be explained by additional cation exchange effects. With the exception of atenolol all chosen model compounds show a high stability towards microbial degradation at aerobic conditions.
All experiments were repeated at least three times at identical conditions, whereby a good reproducibility was observed. Further experiments are currently performed to characterize pH-depending change of sediment surfaces and to investigate the competitive influence of other presented cations.