Long-term batch study of sorption, transformation and extractability to characterize the fate of the veterinary antibiotic sulfadiazine

Stephan Sittig, Roy Kasteel, Joost Groeneweg, and Harry Vereecken
Agrosphere (ICG-4), Forschungszentrum Jülich, Leo-Brandt-Str., 52428 Jülich (Germany)

The occurrence of veterinary antibiotic substances in various environmental compartments is of growing concern. Once released into the environment (e.g. via manure), these organic substances can cause changes in the composition of microbial populations, provoke the development and spreading of resistance genes and finally reach the food chain. The substance under study is the veterinary antibiotic sulfadiazine (SDZ), which belongs to the chemical group of the sulfonamides. These compounds are widely applied in animal husbandry. There are hardly any studies on the macroscopic sorption and desorption behaviour in combination with transformation processes, particularly investigating the sorbed fraction. We are conducting long-term batch sorption experiments to characterize the partitioning between the liquid and the solid phases as well as formation of transformation products. A sequential extraction procedure enables us to analyse the composition of the various sorbed fractions. We applied 14C-labelled SDZ in aqueous solution to fresh soil, originating from an agricultural field (silty loam). Adsorption and desorption studies are conducted for the duration of 60 d and 80 d, respectively. Unique setups for single time-steps allow us to trace the development of the partition process between the liquid and the solid phase and also partitioning within the solid phase. The composition of these liquid phases concerning the parent substance and the transformation products is analyzed. Using Radio-HPLC we find at least five transformation products: 4-hydroxy-sulfadiazine (4-OH-SDZ), 4-(2-iminopyrimidin-1(2H)-yl)-aniline (An-SDZ) and additionally three yet unknown products. By means of a sequential extraction, differently strong bound fractions of the compound can be distinguished. Extractions consist of a mild method (0.01 M CaCl2-solution; 24 h) followed by a methanol extraction (4 h). Finally, a residual fraction is gained by microwave extraction at an elevated temperature (150°C) and pressure (mixture of water and acetonitril, 4:1). Bound residues are determined by combustion. The course of the kinetic adsorption/desorption processes as well as the partitioning of the compound over the various solid phase fractions is observed. Sorption is time-dependent and strongly non-linear. The topsoil shows a significantly higher sorption affinity than the subsoil. While the amount of radioactivity sorbed to the soil matrix increases with time, the extractability decreases significantly, i.e. at the end of the experimental time there is no yield with mild extraction methods. On the contrary, after 60 d, there is still a considerably mass gained with the microwave extraction. Desorption is very slow due to hysteresis. In the topsoil transformation occurs with higher rates, leading to more detectable transformation products as in the subsoil. With our experimental setup it will be possible to set up a kinetic modell for the partitioning of the solute between the liquid and the solid phase. This description will also include an estimation of the transformation parameters.