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How representative are dissipation kinetics determined by sediment-water tests? A case study with pharmaceuticals

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Sediment-water-tests are an established method to predict the behaviour of (micro-) pollutants such as pharmaceuticals in the aquatic environment. Not least due to the diversity of aquatic ecosystems, no standardized test sediment but many different types of sediment are used. Hence it is important to evaluate the representativeness of the test results (e.g., dissipation half-life-times). To address this issue, we investigated selected pharmaceuticals with one test-setup using different sediments from the rivers Roter Main (Germany) and Säva Brook (Sweden).

Four sediments sample were taken from the Roter Main: two sandy batches upstream of Bayreuth (at an interval of one year) and one sandy and one clayey batch downstream the local sewage treatment plant. From the Säva Brook, which does not receive regular input of pharmaceuticals, a clayey batch was tested. To exclude other variables than the sediment, we used synthetic river water containing a carbon source and trace nutrients. 200g of wet sediment and 600mL of synthetic river water were incubated in glass bottles at 15°C in darkness. The water-phase was continuously aerated with pressurized air. After one week of equilibration the supernatant was replaced by fresh synthetic river water containing the pharmaceuticals at an initial concentration of 100 or 200 μg L-1. Moreover, one sediment batch of each river was tested under sterile conditions to assess the influence of sorption. Samples were analysed with an UPLC-QTOF-MS instrument without any sample pretreatment other than filtration. Based on the measured concentrations, we calculated dissipation times (dt50) reflecting the time until 50% of the respective pharmaceutical dissipated from the water phase.

In all non-sterile experiments the concentrations of the pharmaceuticals declined over time with dt50 values between 3 and 55 days. While the beta-blockers metoprolol and propranolol dissipated from the aqueous phase due to sorption and degradation, the acidic pharmaceuticals ibuprofen, furosemide, and naproxen were predominantly eliminated by microbial degradation. In general, the behaviour of ibuprofen and furosemide was similar, with much shorter dt50 in the Roter Main sediments compared to the Säva Brook sediments. In contrast, the dissipation of naproxen and the two beta-blockers proceeded at a similar rate in both sediments. Differences in dissipation kinetics in sediment sampled from the same site in the Roter Main river with a time lag of one year were generally <45 %. The dependence of the dissipation kinetics on the regular exposure of the sediment to pharmaceuticals was low: in sandy sediment sampled upstream and downstream of the sewage treatment plant, the dissipation rates of all compounds were almost identical. The effect of sediment texture (clay vs. sand) on dissipation kinetics was very pronounced for some compounds (propranolol, naproxen), while it was comparatively little for ibuprofen, furosemide, and metoprolol.

Overall, the results of our sediment-water tests do not show a consistent pattern for the tested compounds and sediments. There are compounds for which the dissipation kinetics is drastically different in sediments from different rivers, while for others the sampling location within the individual river seems to have a much larger influence. Based on the large and non-systematic variability of results, we suggest to use more than two sediments if such sediment-water tests are used to determine kinetic parameters for environmental risk assessment and modeling. Moreover, we suggest to benchmark dissipation kinetics of new chemicals by using a broad spectrum of reference substances with known dissipation behaviour. Finally, the mass transport in these test-systems is dominated by diffusive exchange of solutes between water and sediment, while in running waters this exchange is predominantly achieved by advective processes. Therefore, we suggest using recirculating column systems to derive more meaningful degradation rates in river sediments.