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Long-term trends in dissolved iron and DOC concentration linked to nitrate depletion in riparian soils

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The instream concentrations of dissolved organic carbon (DOC) are rising in many catchments of the northern hemisphere. Elevated concentrations of DOC, mainly in the form of colored humic components, increase efforts and costs of drinking water purification. In this study, we evaluated a long-term dataset of 110 catchments draining into German drinking water reservoirs in order to assess sources of DOC and drivers of a potential long-term change. The average DOC concentrations across the wide range of different catchments were found to be well explained by the catchment's topographic wetness index. Higher wetness indices were connected to higher average DOC concentrations, which implies that catchments with shallow topography and pronounced riparian wetlands mobilize more DOC. Overall, 37% of the investigated catchments showed a significant long-term increase in DOC concentrations, while 22% exhibited significant negative trends. Moreover, we found that increasing trends in DOC were positively correlated to trends in dissolved iron concentrations at pH$\leq$6 due to remobilization of DOC previously sorbed to iron minerals. Both, increasing trends in DOC and dissolve iron were found to be connected to decreasing trends and low concentrations of nitrate (below $\sim$6 mg/L). This was especially observed in forested catchments where atmospheric N-depositions were the major source for nitrate availability. In these catchments, we also found long-term increases of phosphate concentrations. Therefore, we argue that dissolved iron, DOC and phosphate were jointly released under iron-reducing conditions when nitrate as a competing electron acceptor was too low in concentrations to prevent the microbial iron reduction. In contrast, we could not explain the observed increasing trends in DOC, iron and phosphate concentrations by the long-term trends of pH, sulfate or precipitation. Altogether this study gives strong evidence that both, source and long-term increases in DOC are primarily controlled by riparian wetland soils within the catchments. Here, the achievement of a long-term reduction in nitrogen deposition may in turn lead to a more pronounced iron reduction and a subsequent release of DOC and other iron-bound substances such as phosphate.