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Disentangling the impact of catchment heterogeneity in a meso-scale catchment on nitrate export dynamics across time scales

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High nitrate concentrations in groundwater and surface water are a long-known but still widespread problem. To most efficiently reduce nitrate pollution, a detailed understanding of catchment organization and the catchment internal processes that drive nitrate mobilization, transport and storage across time scales is needed. Especially in mesoscale catchments (10^1 – 10^3 km^2), spatial heterogeneity adds another layer of complexity to these processes compared to headwater catchments. To address this issue, we analyzed seasonal long-term trends (1983 – 2016) and high frequency event dynamics (2010 – 2016) of nitrate concentrations, loads and the concentration-discharge relationship (CQ-slope) in three nested catchments within the Selke catchment (Germany). Transit time distributions (TTDs) were calculated for each nested catchment to analyze the response of nitrate export to changes in nitrogen surplus. The upper part of the Selke catchment is dominated by forests with only little agriculture and an overall lower nitrogen surplus, while the lower Selke is dominated by agriculture and a higher nitrogen surplus. Surprisingly, we found a disproportionally high contribution to nitrate loads from the forest-dominated upper Selke (64% of average annual load at the Selke outlet), caused by high nitrate concentrations during wet seasons (average of 2.5 mg-N L^{-1} during winter and spring) while dry season nitrate concentrations are relatively low (average of 1.1 mg-N L^{-1} during summer and autumn). These seasonally high concentrations can be explained by the sub-catchment characteristics such as shallow soils and steeper slopes that lead to a low retention capacity and short effective transit times (peak of TTD after 2 years, indicating a fast response to changes in nitrogen surplus). The increase of nitrate concentrations with discharge resulted in a positive CQ-slope that was consistently observed in long-term dynamics and during events. In the lower Selke, nitrate concentrations are relatively constant across seasons (around 3.1 mg-N L^{-1}). This dynamic is caused by deeper aquifers, long effective transit times (peak of TTD at the Selke outlet after 14 years, indicating a delayed response to changes in nitrogen surplus) and legacy stores of nitrate that constantly release into the Selke River. Consequently, the lower Selke dominates nitrate concentrations and loads exported during dry seasons and is characterized by lower CQ-slopes compared to the upper Selke. Our study shows that the contribution of different sub-catchments to elevated nitrate concentrations can vary greatly between seasons, flow conditions and in their response to changes in nitrogen surplus. It is, therefore, not enough to focus on areas of highest nitrogen surplus – such as the upper Selke; instead, an assessment of all characteristic sub-catchments, their temporally variable contribution to nitrate export and their specific TTDs is

needed to place reduction measures most effectively and to estimate realistic time scales for their success.