A multi-evidence approach is presented to examine the influence of ‘small-scale’ and ‘large-scale’ spatial processes on hydrologic functioning in two nearby watersheds in the Catskill Mountains, New York State, USA. The Neversink (176.0 km²) is a steep, mountainous watershed cut by two distinct branches. In contrast, Townbrook watershed (36.8 km²) has much thicker till deposits and less topographic relief. Nested sampling techniques are used to examine changes in water chemistry, discharge, and estimated residence time (using isotope tracers) across spatial scale. In both watersheds, small-scale ‘hillslope’ processes appear to have significant effects on watershed functioning below 2-3 km². Groundwater springs are responsible for saturation patterns and hydrologic connectivity in a study sub-watershed (3.2 km²) in Townbrook. End-member mixing analysis (EMMA) indicates that saturation-excess overland flow from spring-caused saturated areas may be a significant ‘quick’ runoff source (>40% of volume) during precipitation events. The EMMA also suggests that at scales below 2-3 km² low-flow chemistry and discharge is dominated upland groundwater springs in Townbrook. However, synoptic sampling in both watersheds reveals a reduction in variability at larger scales (>3 km²) that is likely due to ‘emergent’ large-scale processes dominating hydrologic response. In the Neversink, spatial patterns of alluvial deposits appear to control summer baseflow discharge and chemistry. Furthermore, the proximity and connectivity to steep, upland hillslopes may control the relative influence of small and large scale processes in Townbrook. Overall, the results suggest geomorphology and physiographic position must be accounted for in spatially structured landscapes. The study presents a novel watershed conceptualization that identifies repeatable hydrologic landscape units and captures spatial variability without the need to explicitly represent all the small-scale process heterogeneity. Therefore, the results of this study could improve process conceptualization and spatial modeling in similar complex landscapes.