

## Exploring Nitrogen Legacies and Time Lags in Anthropogenic Landscapes: A 200-Year Longitudinal Study of the Mississippi and Susquehanna Watersheds

Kimberly Van Meter, Nandita Basu, and Philippe Van Cappellen

University of Waterloo, Earth and Environmental Sciences, Waterloo, Canada (nandita.basu@uwaterloo.ca)

Global flows of reactive nitrogen (N) have increased significantly over the last century in response to land-use change, agricultural intensification and elevated levels of atmospheric N. Despite widespread implementation of a range of conservation measures to mitigate the impacts of N-intensive agriculture, N concentrations in surface waters are in many cases remaining steady or continuing to increase. Such time lags to the recovery of surface water quality are increasingly being attributed to the presence of legacy N stores in subsurface reservoirs. It has remained unclear, however, what the magnitudes of such stores might be, and how they are partitioned between soil and groundwater reservoirs. In the present work, we have synthesized data from numerous sources to develop a comprehensive, 200-year dataset of N inputs to the land surface of the continental United States. We have concurrently developed a parsimonious, process-based model that utilizes this N input trajectory together with a travel time-based approach to simulate biogeochemical transformations of N along subsurface pathways. Model results allow us predict the magnitudes of legacy N in soil and groundwater pools and to predict long-term N-loading trajectories over the last century and into the future. We have applied this modeling approach to two major U.S. watersheds, the Mississippi River and Susquehanna River Basins, which are the sources of significant nutrient contamination to the Gulf of Mexico and Chesapeake Bay, respectively. Our results show significant N loading above baseline levels in both watersheds before the widespread use of commercial N fertilizers, largely due to 19th-century conversion of natural forest and grassland areas to row-crop agriculture, although the temporal patterns of this loading differed between the two watersheds due to differences in the trajectories of land-use change. Using the model, we estimate spatiotemporal patterns of N accumulation in both groundwater and soil organic matter in response to increases in N inputs to agricultural soil as well as changes in N residence times across the terrestrial system. Simulations of future scenarios allow us to predict changes in N-loading as determined by both ongoing inputs and the existence of biogeochemical and hydrologic N legacies.