Advancing Methods for Contaminant-source Attribution in Complex Aquifer Settings: Applications in the Northwestern Appalachian Basin, USA

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Groundwater, which serves as a major source of drinking water globally, is susceptible to contamination from a variety of natural and anthropogenic sources. Elucidating sources of contamination and the processes controlling contaminant occurrence in groundwater is critical to informing remediation activities and limiting public exposures. However, contaminant-source attribution is challenging in aquifers characterized by complex structure and stratigraphy or where multiple contaminant sources co-occur. To help address this issue, we advance an approach combining multivariate statistical clustering techniques, geochemical modeling, geospatial analysis, and noble gas geochemistry to support inferences on factors controlling groundwater quality. We apply our integrated approach to measurements of major ions, trace metals, noble gases, and per-and polyfluoroalkyl substances (PFAS) collected from 216 domestic groundwater wells in southeastern Ohio and northern West Virginia. Within this region, groundwater quality is threatened by potentially polluting extractive industries (e.g. coal mining, oil and gas development) and geogenic contaminants that occur within a complex, perched-aquifer setting. Through hierarchical cluster analysis, we classify the well-water samples into nine distinct clusters of similar major ion and trace-metal composition. Clusters representing recently recharged groundwaters, primarily located in upland regions, appear susceptible to surface-related impacts, such as nitrate pollution from agricultural activities, salinization from domestic sewage and road-salt applications, and PFAS contamination from atmospheric or industrial sources. Domestic wells situated in valley settings primarily intercept long-residence-time groundwaters with occasional influences from natural basin brines. Arsenic impairments, likely from naturally occurring geologic sources, were associated with alkaline groundwaters with major-ion compositions altered by ion exchange or with anoxic, metals-rich groundwaters. The chemistry of well-waters collected near active and abandoned coal mines was influenced by mixing with as much as 4-10% mine-pool discharge. Our approach successfully delineates groundwaters impacted by varied anthropogenic and geogenic sources and has implications for contaminant-source attribution efforts in complex aquifer settings globally.