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Regional-scale redox controls on the contamination of shallow groundwater with nitrate, manganese, arsenic, iron and methane in Alberta, Canada

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Providing high-quality drinking and agricultural water is key to supporting an increasing global population, but contamination of shallow aquifers remains a widespread challenge in many countries. We investigated the influence of redox conditions on the occurrence and fate of nitrate, manganese, arsenic, iron and methane in shallow groundwater on a province-wide scale in Alberta, Canada. The objective was to document how redox zonation in shallow aquifers governs the occurrence and fate of groundwater contaminants using a combination of geochemical, isotopic and microbiological approaches.

Aqueous geochemistry data from >143,000 groundwater samples collected between 1960 and 2024 from domestic and monitoring wells completed in shallow aquifers (< 150 m) revealed that only 3 % of the samples exceeded the Canadian maximum acceptable concentration (MAC) for nitrate in drinking water (10 mg/L NO_3^--N), with exceedances mainly in wells shallower than 30 m. At greater depths, redox conditions were found to be frequently suitable for denitrification, resulting in nitrate removal from groundwater. These redox conditions, however, promoted Mn(IV) reduction in the upper post-oxic redox zone, leading to elevated Mn^{2+}concentrations that exceeded the Canadian MAC of 0.12 mg/L in some groundwater samples. Aquifers with groundwater in the lower post-oxic redox zone displayed elevated concentrations of Fe^{2+} (> 1.0 mg/L), and locally elevated As concentrations, likely due to reductive dissolution of iron-oxides, where As- and Fe-containing aquifer materials exist. In more reducing environments, considerable portions of shallow groundwater were found to have sulfidic and methanic conditions. While H_2S generated by bacterial sulfate reduction rapidly precipitates as iron sulfide minerals due to often elevated dissolved Fe^{2+} concentrations, occurrence of microbial methane was found to be widespread in shallow groundwater in Alberta, Canada. This methane may contribute to greenhouse gas emissions where groundwater is produced or is subject to groundwater – surface water interactions. This study revealed that the combination of geochemical, isotopic and microbiological approaches provides a powerful tool to identify redox controls on the occurrence and fate of groundwater contaminants such as nitrate, manganese, arsenic and methane even on large spatial scales.