

Causes for decadal-scale patterns of nitrate in surface waters and shallow groundwater in large watersheds of Western Canada

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Providing high-quality drinking, agricultural and recreational water is of key importance for supporting an increasing global population, but nitrate contamination of surface waters and shallow aquifers is a widespread problem in many countries. In this study, we investigated the patterns of nitrate occurrence in surface waters and groundwaters throughout the last few decades for several large watersheds in Alberta, Canada. Key objectives included the assessment of temporal patterns of nitrate occurrence in surface waters and shallow groundwater, and the determination of the sources and the fate of nitrate in the investigated aqueous systems using a combination of geochemical, isotopic and microbiological approaches.

Average annual nitrate concentrations during baseflow for surface water sampling sites were typically < 0.2 mg/L NO₃⁻-N at upper reaches and up to 1.5 mg/L NO₃⁻-N at lower reaches of main rivers with either constant or slightly increasing trends between 1978 and 2024. Isotopic analyses of surface water samples (d¹⁵N and d¹⁸O of nitrate and d¹¹B values) revealed that nitrate concentration peaks during snowmelt are often caused by mineralization of soil organic nitrogen and nitrification of ammonium, while elevated nitrate in downstream regions is both derived from effluents of wastewater treatment plants and agricultural return flows. Aqueous geochemistry data from >130,000 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 exceed the maximum allowable concentration for drinking water in Canada of 10 mg/L NO₃⁻-N. Time series analysis, however, showed that in 17% of the townships nitrate concentrations in shallow groundwater have significantly increased throughout the last decades. The isotopic composition of nitrate revealed that elevated nitrate concentrations in groundwater are frequently the result of cattle manure applications. Chemical, isotopic and metagenomic analyses also provided evidence that denitrification is an important nitrate removal process in some aquifers. This study revealed that the combination of geochemical, isotopic and microbiological approaches is a powerful tool to investigate the occurrence, origin and fate of nitrate in surface water and shallow groundwater systems of large watersheds.