## Boron Isotope-Based Assessment of Human Activity-Induced Contamination in the Ryukyu Limestone Groundwater System

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Boron isotopic ratio ( $\delta^{11}$ B) is a potential tracer for identifying contaminants in environmental systems such as rivers and groundwater. In this study, we developed a simplified separation method for boron isotopes and validated its accuracy using well-characterized reference materials (RMs). This research presents the first published boron isotopic compositions of Japan-made certified reference materials (CRMs) for river water (NMIJ CRM 7202-c:  $-8.81 \pm 0.19\%$ , JSAC 0301-4a:  $14.33 \pm 0.49\%$ ) and tap water (NMIJ CRM 7203-a:  $17.86 \pm 0.28\%$ ).

Furthermore, we applied the developed method to quantitatively assess contamination from agricultural activities in the Quaternary limestone aquifers of southern Okinawa Island, SW Japan. Boron concentrations and  $\delta^{11}B$  values were measured in groundwater, spring water, soil leachates, and potential contaminant sources collected between August 2020 and August 2021. A positive correlation between boron and nitrate-nitrogen concentrations in groundwater suggests co-migration, indicating that boron serves as a valuable tracer in the Ryukyu Limestone region. Monthly groundwater monitoring revealed boron concentrations ranging from 12 to 168 ppb, with  $\delta^{11}B$  values between 19 and 38 ‰.

This study identified four distinct endmembers with different boron characteristics, providing insights into contamination sources and processes through mixing models. Our calculations indicate that precipitation, containing less than 1% seawater component, is the primary recharge source for groundwater in this region. The estimated contributions of major contaminants, including manure and chemical fertilizers, were less than 1% in groundwater and approximately 3% in soil leachates.

Overall, this study demonstrates the effectiveness of boron isotopes as a quantitative tracer for assessing fertilizer-induced contamination in the Ryukyu limestone watershed, offering a valuable tool for environmental monitoring and groundwater management.

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