

## Importance of surface geology in regulating As concentration of alluvial groundwater

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Arsenic concentrations in the groundwater of alluvial plains generally show a high degree of spatial variability. This study geochemically explores the reason for the spatial variation of As concentration by investigating the processes and the sources of chemicals that affect As concentrations. This study was performed in the point bar area of Mankyeong River watershed, which is mainly used for crop (rice) cultivation like other alluvial plains in the world, by inspecting the vertical changes in hydrogeochemistry and geology at 7 sites. All the sites showed As concentrations increasing with depth within the shallow depth range and decreasing at the greater depths, the vertical pattern typically observed in the alluvial aquifers where the reductive dissolution of Fe-(hydr)oxides is the major As release mechanism. The geochemical data show that  $\text{NO}_3$  and  $\text{SO}_4$  originating from the paddy field play important roles in suppressing the increase of As concentrations within the shallow and the deep depth ranges, respectively. Furthermore, the supply of those chemicals to the aquifer is strongly dependent on the geologic condition of the aquifer. In the sites where the aquifer is covered with thick silt layer (~ 5 m), the supply of  $\text{NO}_3$  and  $\text{SO}_4$  is largely limited; as a result, the maximum As concentrations in the vertical profile were 3 to 5 times higher than those at other sites, whose land surfaces were permeable. This result indicates that the surface geology could be the important indicator for the As concentration in the alluvial groundwaters.

## $\delta^{44}\text{Ca}$ traces chemical weathering of hydrothermal calcite in the Southern Alps of New Zealand

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The marine Ca isotope record has emerged as a powerful tool for understanding chemical exchanges between the continents, oceans, and atmosphere. However, interpretation of the record remains uncertain because factors governing isotopic variation of Ca during chemical weathering and riverine transport are not well understood.

We collected 56 samples from 42 rivers, including glacial outlets, draining the Southern Alps of New Zealand and used a MC-TIMS to analyze  $\delta^{44}\text{Ca}$  ( $^{44}\text{Ca}/^{40}\text{Ca}$  relative to seawater). The Southern Alps have a nearly uniform bedrock chemistry but large lithologic, climatic, and tectonic gradients. East of the main divide, rivers drain greywacke or schist and experience relatively low precipitation and uplift rates. West of the divide, rivers drain schist and experience relatively high precipitation and uplift rates. Schist and greywacke contain trace hydrothermal calcite (~3 vs. <1%, respectively).

Glacial and non-glacial rivers draining greywacke display  $\delta^{44}\text{Ca} = -1.15$  to  $-1.32$  (avg. =  $-1.24 \pm 0.06$ ; n=8). Rivers draining schist east and west of the main divide display  $\delta^{44}\text{Ca} = -0.92$  to  $-1.09$  (avg. =  $-0.97 \pm 0.06$ ; n=12). Glacial rivers draining schist with  $\delta^{44}\text{Ca} = -0.96$  to  $-1.09$  (avg. =  $-1.02 \pm 0.06$ , n=5) are  $^{40}\text{Ca}$ -enriched compared to non-glacial rivers draining schist with  $\delta^{44}\text{Ca} = -0.92$  to  $-0.96$  (avg. =  $-0.94 \pm 0.02$ ; n=7).

Calcite in schist has  $\delta^{44}\text{Ca} = -0.92$ , whereas the silicate host rock has  $\delta^{44}\text{Ca} = -1.81$ . Waters display two-component mixing between these end-members when the data are examined as  $\delta^{44}\text{Ca}$  vs. Ca/Sr. Calculations suggest calcite contributes ~94 and ~64% of the Ca in rivers draining schist and greywacke, respectively. We attribute  $^{40}\text{Ca}$  enrichment in glacial streams to enhanced release of  $^{40}\text{Ca}$  from silicate minerals by glacial grinding. These data demonstrate that  $\delta^{44}\text{Ca}$  robustly traces carbonate and silicate weathering, and confirm that most riverine Ca emanating from active orogens is not derived from weathering reactions that consume atmospheric  $\text{CO}_2$  over geological time scales.