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Boron Isotope Fractionation in Groundwaters as an Indicator of Permafrost Past Conditions in the Fractured Crystalline Bedrock of the Fennoscandian Shield

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Introduction

We present boron data from the Äspö site groundwaters in Sweden and also from the Palmottu natural analogue site in Finland. During the last million years the Fennoscandian Shield has been subjected to several glaciations, the last of which (Weichselian Ice Age) ended only 10ka ago. It is estimated that the maximum thickness of the continental ice sheet in the central part of the area was about 3km some 20ka ago. Palmottu is characterised by an upper, dynamic groundwater flow system, with typical bicarbonate groundwaters which can be partly explained by mixing processes, but nevertheless show a clear evolutionary trend from Ca-HCO_3 types to deeper Na-HCO_3 types. At greater depths, where low permeable conditions prevail, Na-Cl and Na-SO_4 type groundwaters are present. The coastal location of Äspö makes its hydrochemistry different: only Cl-Ca-Na and Cl-Na-Ca type groundwaters occur at depth. In both sites, there is a sizeable $\delta^{18}O$ depleted groundwater component that may represent glacial melt water. These waters reflect a colder climate recharge which most likely took place during the retreat of the Weichselian ice sheet. In this context, boron isotope ratios are used as tracer of the degree of water-rock interaction and mixing processes in groundwaters. Sixty-eight water samples have been analysed including surface waters, fresh, brackish and saline groundwaters. The chemical analyse of the water samples was performed by means of inductively coupled plasma mass spectrometry for B concentrations and by mass spectrometer for the $\delta^{11}B$.

Chemical and isotopic data

In the investigated hydrosystems, the boron concentration gradually increases with depth from few ppb in surface waters (snow, lake, spring) to several hundred of ppb in the deepest most concentrated waters. In comparison, the Baltic samples have a higher concentration (750ppb<[BBALTIC]<1510ppb) somewhat less, however, than the present mean seawater ([BMSW]=4 ±0.5ppm, Spivack and Edmond, 1987). The $\delta^{11}B$ values of the groundwaters range from 18‰ in the less evolved waters (dilute HCO_3, Ca-HCO_3) to 51.9‰ in a Cl-Na-Ca water-type (Äspö at 452m). The most of the high saline waters have a significantly more positive signature than Baltic seawater ($\delta^{11}B$ = 39.63‰ ±0.5, n=8) or than the present mean ocean seawater (δ^{11}BMSW = 40‰ ±0.5, Barth, 1993). These groundwaters are the most ^{11}B-enriched GW ever found in crystalline shield environments and are rather unexpected because water-rock interactions with granite minerals lead to a ^{11}B depletion (Vengosh, 1992) of the fluid. For example, the deep saline GW and brines reported from the Canadian Shield (-1.7‰<$\delta^{11}B$<35.6‰, Bottomley et al., 1994) never overpass the marine boron isotopic composition. On the other hand, such high $\delta^{11}B$ values (up to 57.4‰) are commonly encountered in marine brines (Vengosh et al., 1991; 1992). During fractional evaporation of sea water a gradual increase in the $\delta^{11}B$ values of the residual brines is observed and correspond to a selective uptake of ^{10}B by the salts (Vengosh et al., 1992). A $\delta^{11}B$ versus B diagram shows a general trend of ^{11}B enrichment with increasing B contents. The Baltic samples, however, suggest that processes controlling the boron isotopic composition of these waters are not, solely, related to the control of salinity. We calculated the mixing lines between different potential end-members in order to better constrain the data. A first mixing line relies the most depleted Ca-Na-HCO_3 GW sample to the deepest Baltic. This evolutionary pathway is likely to reflect the Holocene water cycle through evaporation, precipitation and water rock interaction specific to the Fennoscandian Shield. All water types, but the Na-SO_4 one, are involved in this trend. A second mixing line relies the most enriched Ca-Na-HCO_3 GW sample to the heaviest sample which is of the Cl-Na-Ca type. This evolutionary pathway reflects a fractionation process leading to an ^{11}B enrichment of about 10‰. Altogether, the two evolution trends described by these mixing lines are needed to take into account the whole data set. The fact that poorly evolved water types have also been affected by an ^{11}B enrichment suggests rather freezing processes occurring under permafrost conditions than precipitation of salts due to freezing concentration and evaporation under periglacial conditions.

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