

## How do contact-metamorphic aquifers leak? Evidence from Rb, Sr, and U contents and Sr, Pb, and stable isotope shifts between metachert aquifer and marble aquitard

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Impure limestones with thin horizontally bedded metachert layers have been intruded and contact-metamorphosed by the Bufa del Diente alkali syenite (NE-Mexico). There is mineralogical and stable isotope evidence that infiltration of magmatic fluids into the massive marbles was confined to a 17 m wide zone along the contact of the intrusion. In contrast, influx of fluids into the metachert layers to distances of more than 400 m from the contact is documented by up to 4 cm thick wollastonite rims on both sides of the metachert layer. These rims were produced by the infiltration-driven reaction calcite + quartz = wollastonite + CO<sub>2</sub>. At the marble-ward border of wollastonite rims, there are 0.5 to 1 mm thin meta-argillite rinds consisting mainly of vesuvianite and phlogopite. They are the residue of the original limestone after calcite (and quartz) dissolution. Fluid inclusion studies have shown that the infiltrating magmatic fluid was a high-salinity brine and that CO<sub>2</sub> produced by the wollastonite-forming reaction was immiscible and exsolved as a low density CO<sub>2</sub>-rich H<sub>2</sub>O-CO<sub>2</sub> fluid. Oxygen isotope studies showed that the influx of brine occurred dominantly at the quartz-wollastonite boundary.

We measured Rb, Sr, and U contents and Sr and Pb isotopic compositions across the wollastonite-rimmed metachert layer along a small scale profile (Fig. 1) about 90 m from the contact of the intrusion. The results are compared to earlier analysed  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  profiles from the same sample:

1. The Sr and Pb isotopic signature of the alkali syenite introduced by a magmatic brine into the aquifer can be traced at least to 90 m away from the contact. This is also seen in the oxygen isotope systematics of wollastonite and quartz.

2. Sr and Pb contributions from the alkali syenite can be traced only 3 to 5 cm from the wollastonite-marble boundary into the marbles. This is about the same distance as revealed by C and O isotopic alterations in the hanging-wall marble. Sr and Pb shifts are coherent.

3. The thin meta-argillite rinds are distinctly enriched in Rb, Sr, and U (Fig. 1). Sr in these rinds, as well as Rb at the outer border of the rind, is dominantly residual from the carbonate dissolution. Rb contents at the inner border are distinctly higher, and in part derived from the magmatic fluid. U was also introduced by the highly-saline magmatic fluid.

4. Covariation trends of carbon and oxygen isotopes across the alteration front into the hanging wall marble show that the alteration was caused by the upward-moving immiscible CO<sub>2</sub>-rich fluid, which was produced by the decarbonation reaction.

5. The isotopic variation of Sr, Pb, C, and O across the aquifer-marble profiles suggests advection rather than diffusion of the immiscible CO<sub>2</sub>-rich H<sub>2</sub>O-CO<sub>2</sub> fluid into the overlying marble.

6. U initially transported with the brine partitioned to some extent into the CO<sub>2</sub>-rich fluid and was scavenged by calc-silicate minerals within the thin meta-argillite rind.

7. Radioautographs show that about 20 to 40 times more U is present in the upper meta-argillite rind than in the lower one (Fig. 1). This implies that most of the CO<sub>2</sub>-rich fluid was lost through the upper border of the aquifer. The U distribution corroborates with the C and O isotope profiles in the marbles: above the wollastonite-marble margin, there is a clear covariation between C and O and a wide alteration zone, whereas below the wollastonite-marble margin, there no correlation and only a very small alteration zone.

These observations imply:

The infiltrating highly saline aqueous brine did not leave metachert. The aquifer borders were impermeable with respect to this brine. In situ produced CO<sub>2</sub> unmixed as a CO<sub>2</sub>-rich H<sub>2</sub>O-CO<sub>2</sub> fluid that escaped from the aquifer and infiltrated the overlying marble. The growing wollastonite rim around the metachert acted as a semipermeable membrane that separated the two fluid systems.

The dispersion of trace elements from the aquifer

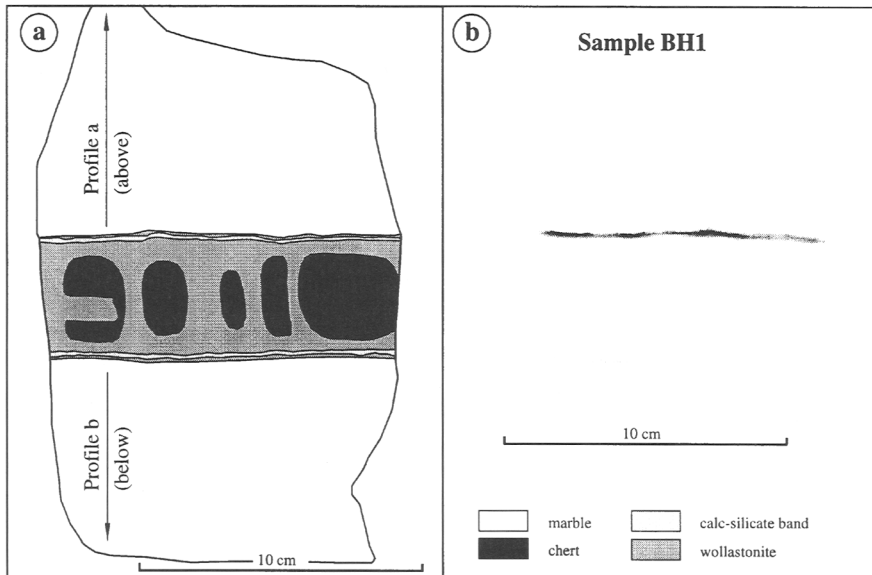


FIG. 1. Drawing of sample BH1, about 90 m from the contact of the intrusion. The calc silicate layer has variable amounts of vesuvianite, phlogopite, albite, plagioclase (An 15–27), Na-rich chlorine scapolite, diopside, titanite, and grandite<sub>ss</sub>. **B.** The radioautograph (redrawn) shows the distribution of U. The meta-argillite bands, left behind after the dissolution of impure marble, show distinct radiation-induced blackening.

into the marble (and vice versa) depends on the complex interplay of (1) availability, (2) mobility, (3) partitioning, and (4) scavenging. Availability is controlled by the amounts present in the marble and the metachert and the amount of trace elements introduced by magmatic fluid, which is governed by the brine-rock exchange behaviour along the chromatographic column of the aquifer. Mobility is controlled by element specific solubilities and the amount of fluid available, which corresponds to the amount of CO<sub>2</sub> produced that can unmix and escape from the aquifer. As only elements dissolved in the

CO<sub>2</sub>-rich fluid can infiltrate into the marble, the dispersion pattern depends strongly on the partitioning behaviour of different elements between brine and CO<sub>2</sub>-rich fluid and the partitioning behaviour between CO<sub>2</sub>-rich fluid and the infiltrated marble. Scavenging layers, such as the meta-argillite rinds with respect to Rb and U, may severely deplete the fluid in certain trace elements. The band at the upper margin of the aquifer is wider and darker than the one at the lower contact. U contents of the marble, chert, and wollastonite-altered aquifer are below detection limit.