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Large negative $\delta^{238}U$ anomalies in endogenic-type travertine systems

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ABSTRACT

We report exceptionally negative $\delta^{238}U$ values for spring water (−2.5‰ to −0.8‰) and travertine calcite (−3.2‰ to −1.1‰) from an area where the Jemez lineament intersects the western margins of the Rio Grande rift, west-central New Mexico (southwestern United States). The highest anomalies come from the southern margins of the Valles Caldera and are related to upwelling CO_2-charged spring water forming travertine mounds along joints and faults. The anomaly likely occurs due to CO_2 lixiviation of uranium in a deep-seated reduced environment where ^{235}U is preferentially leached along a long flow path through Precambrian granitic basement, resulting in spring water with exceptionally low $\delta^{238}U$ values inherited by the calcite that precipitated near or at the surface at relatively low temperatures, i.e., ~40 °C (modern temperatures). The lowest $\delta^{238}U$ values are preserved in settings where upwelling waters are least diluted by oxidized aquifer groundwaters. Given these low $\delta^{238}U$ values in travertine are associated with and possibly indicators of upwelling CO_2 related to tectonic and magmatic activity, studies such as ours may be used to identify this association far back in time.

INTRODUCTION

Uranium (U) was once thought to be too heavy to undergo measurable natural mass-dependent isotope fractionation such that the $^{238}U/^{235}U$ value in nature was thought to be invariant at 137.88. Instead, improved instrumentation and methods show otherwise (Hiess et al., 2012; Stirling et al., 2007). An extreme variant of this atomic ratio was measured as 130.17 in calcium-aluminum-rich inclusions in meteorite Curious Marie (Tissot et al., 2016). The $^{238}U/^{235}U$ for natural materials on Earth has a surprisingly large variation as well. Expressed as $\delta^{238}U$ with respect to a newly reported bulk earth value of $^{238}U/^{235}U = 137.818$ (Hiess et al., 2012), this spread exceeds 8‰. Except for a few mantle-assigned samples of zircon and titanite, the variation of $\delta^{238}U$ on Earth is roughly 5.0‰ (Hiess et al., 2012; Murphy et al., 2014). This per-mil-level spread is significant to the accuracy of U-Pb and Pb-Pb dating. In addition, it is potentially a useful geochemical tracer for igneous and low-temperature aqueous geochemistry studies (Moynier et al., 2013). Carbonate rocks, including limestones, corals, and speleothems, show a large spread in values (~1.3‰; Hiess et al., 2012), and few results exist for travertine.

The natural U isotope system is measured as two ratios, $^{234}U/^{238}U$ and $^{238}U/^{235}U$, where the $^{234}U/^{238}U$ activity ratio is calculated as $\delta^{234}U = \{[(^{234}U/^{238}U)_{measured} / (^{234}U/^{238})_{secular\ equilibrium})] - 1\}*1000$ and has large variance (thousands of ‰) due to the vulnerability of ^{234}U (e.g., seated in damaged crystal lattices) to leaching (Chabaux et al., 2003). However, supporting a 5‰ spread in $\delta^{238}U = \{[(^{238}U/^{235}U)_{measured} / (^{238}U/^{235}U)_{standard\ NBL112A}] - 1\}*1000$ is more difficult and requires explanations related to nuclear processes of U and other heavy elements (Bigeleisen, 1996; Moynier et al., 2013; Van Hook, 2011). See Supplemental Material[1] regarding standard NBL-112A. Field and experimental results show that $\delta^{238}U$ is sensitive to marine and/or terrestrial redox events (Brennecka et al., 2010; Murphy et al., 2014), adsorption (Brennecka et al., 2011), microbial processes (Bopp et al., 2010), and leaching (Stirling et al., 2007). More specifically, these processes affecting the $\delta^{238}U$ value relate to nuclear field shift effects of the U atom (Bigeleisen, 1996).

Many New Mexico (southwestern United States) travertine deposits have apparently formed from upwelling fluids containing deeply sourced CO_2 and magma- and/or mantle-derived He as shown by $^3He/^4He$ analyses of travertine-depositing springs (Crossey et al., 2016; Newell et al., 2005). These deposits are commonly aligned along fault zones and/or associated with magma bodies and form mounds fed by spring vents (Crossey et al., 2016). In these upwelling systems, two possible settings for low $\delta^{238}U$ values (i.e., <1‰) could be derived: (1) in the near surface via reduction-oxidation by biological activity (Bopp et al., 2010) or by mineralization (Murphy et al., 2014) during travertine precipitation, or (2) in the deeper subsurface from leaching (Hiess et al., 2012) along groundwater flow paths or from deep microbial activity. Shallow reduction of ^{238}U relative to ^{235}U at or near the surface versus deeper below the surface is testable by analyzing both the water and travertine. Potentially, with the deep-sourced scenarios, the $\delta^{238}U$ values in calcite can be used to identify a connection with deep-seated solutions and/or gases millions of years back in time. Examples of old travertine deposits include Proterozoic geothermal travertine in northern Canada (Rainbird et al., 2006); the Upper Jurassic travertine deposits in the Deseado Massif of Patagonia, Argentina (Guido and Campbell, 2018); the "fossil" upper Miocene to lower Pliocene travertine deposits in the lower Bouse Formation in Arizona, United States (Crossey et al., 2017); and geothermal travertine-depositing water in the Quaternary Jemez volcanic system of New Mexico, where travertine deposition extends

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[1]Supplemental Material. Supplemental text including methods, Figures S1, S2, and S3, and Tables S1 and S2. Please visit https://doi.org/10.1130/GEOL.S.23881170 to access the supplemental material, and contact editing@geosociety.org with any questions.

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back past the ~650 k.y. range of U-series dating (Ricketts et al., 2014) and perhaps for the millions of years of the volcanic geothermal system (Goff and Shevenell, 1987). From this latter area, we analyzed both water and travertine and report anomalously low values of $\delta^{238}U$ in both.

SAMPLES AND GEOLOGIC SETTING

Travertine deposits from two general areas in New Mexico, (1) the Mesa del Oro and Belen travertine sites and (2) travertine mounds along the Jemez and Nacimiento fault zones (Tierra Amarilla anticline aka the San Ysidro anticline, Jemez Springs, Soda Dam, and other springs along the Nacimiento fault zone), all located along the western edge of the Rio Grande rift in west-central New Mexico (Fig. 1), are the geologic settings pertinent to this research. The Mesa del Oro and Belen sites are near the northern margin of the Socorro magma body, while Soda Dam, Jemez Springs, and Tierra Amarilla anticline are on the southern margin of the Valles Caldera in the Jemez Mountains (Ricketts et al., 2014). Five samples of travertine for this study come from Mesa del Oro, which is a large, thick sequence of travertine that overlies Mesozoic and Paleozoic sedimentary rocks (Forbes, 1994). At the Belen site, two samples of travertine come from the Temple Cream and Scheherazade quarries owned by New Mexico Travertine, Inc., where the travertine has formed on Paleozoic rocks (Ricketts et al., 2014). Most of our travertine and water samples are from Soda Dam and Tierra Amarilla anticline, where the most anomalous $\delta^{238}U$ values were measured. The Soda Dam and Jemez Springs geologic settings are travertine deposits from hot-spring vents located along the Soda Dam fault near the unconformity between Precambrian granitic basement and an overlying thin sequence (<200 m) of Paleozoic carbonate rocks (Goff and Kron, 1980). The Tierra Amarilla anticline geologic setting is travertine capping Jurassic and Triassic clastic sediment and probably an overall thick sequence of Paleozoic rocks deeper in the subsurface. We analyzed several spring-water samples from the study area and from other spring sites extending from southern New Mexico into central Colorado (Fig. 1).

THE URANIUM ISOTOPE ANOMALY

The spring-water and travertine $\delta^{238}U$ values for Soda Dam, Jemez Springs, Peñasco Springs, and Tierra Amarilla anticline range from $\delta^{238}U_{travertine} = -3.24‰ \pm 0.16‰$ to $-1.10‰ \pm 0.03‰$ and $\delta^{238}U_{water} = -2.85‰ \pm 0.34‰$ to $-0.83‰ \pm 0.06‰$, extending the known variance in carbonate rocks by ~2.0‰ (Fig. 2). Methods for this measurement are described in the Supplemental Material. Spring water near the Soda Dam site yielded values that range from $-2.47‰ \pm 0.18‰$ to $-1.09‰ \pm 0.29‰$, showing that the travertine is inheriting this anomaly from endogenic spring water and that the values are not the result of shallow-level carbonate precipitation fractionation. Other spring-water and travertine values from a larger region (Colorado and New Mexico) exhibited little or no anomaly, which indicates that the study area, centered

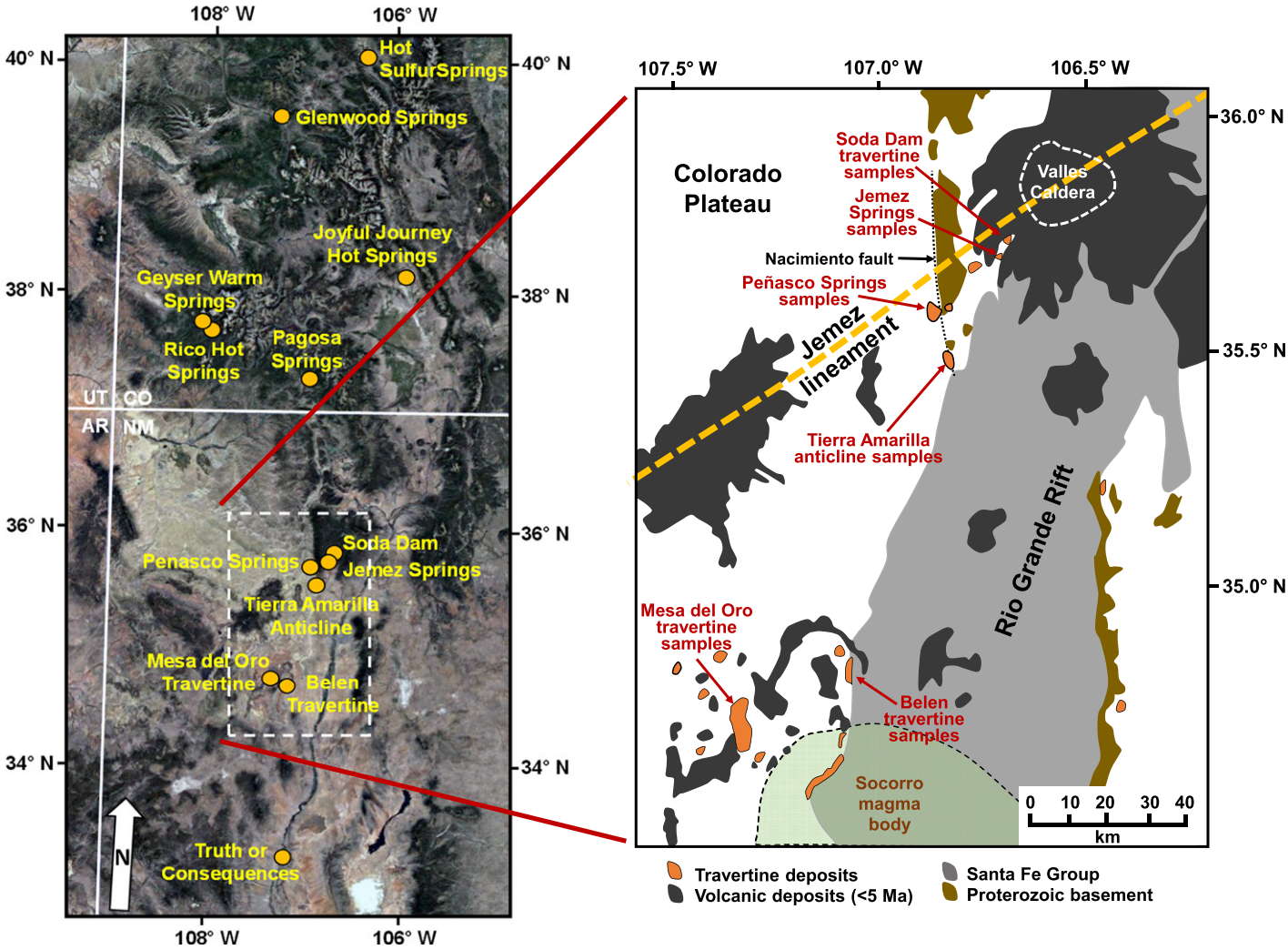

Figure 1. Sample locations of thermal springs and associated travertine along the Rio Grande rift in New Mexico and Colorado (southwestern United States) that were sampled for this study. Inset shows area of primary study. Figure inset was modified after Newell and Crossey (2005).

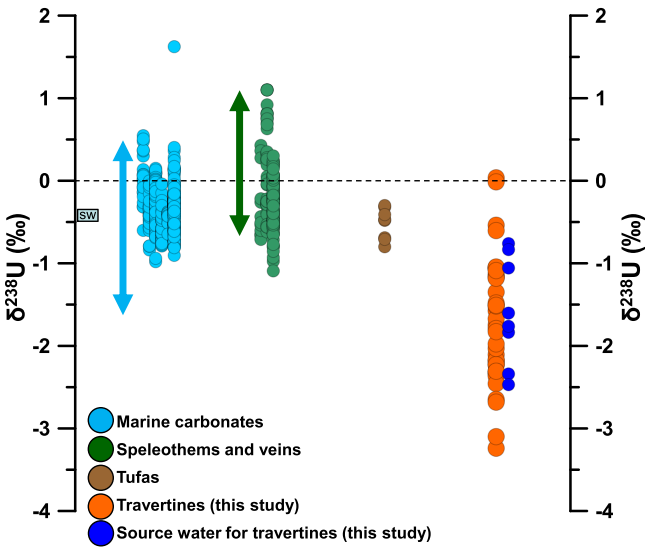

Figure 2. Global $\delta^{238}U$ signatures for carbonate rocks. Marine carbonates (light blue) include limestones, dolostones, and corals; values from Bartlett et al. (2018), Brennecka et al. (2011), Elrick et al. (2017), Hiess et al. (2012), and Jost et al. (2017). Speleothem and vein calcite and/or aragonite (green) are from Cheng et al. (2013), Hiess et al. (2012), and this study. Values for tufas (brown) are from Rovan et al. (2021). Travertines (orange) and their source waters (deep blue) are from this study. Double arrows are global ranges for marine carbonates (blue) and speleothems and veins (green) reported by Andersen et al. (2017). Seawater (SW) value is shown in small box. See Tables S1 and S2 (text footnote 1).

around the broader Valles Caldera geothermal system (McGibbon et al., 2018), is the only place where the anomaly has been distinctly measured thus far.

Crossey et al. (2009) used helium isotope and carbon isotope values to show that a small but significant percent of the helium and its CO_2 carrier gas within or associated with spring waters have mantle isotope signatures, and therefore some of the gas and/or fluid is coming from the mantle. Our exceptionally low $\delta^{238}U$ values come from analyses of spring water and travertine calcite surrounding the Valles Caldera at the western margin of the Rio Grande rift in an area that exhibits strong evidence for magma- and/or mantle-derived He (Crossey et al., 2016; Goff and Janik, 2002). These samples range in age from recent to a few million years (Ricketts et al., 2014). Periods of travertine deposition are likely linked to deep-seated or long-path carbonic spring activity along faults linked to local tectonic and/or magmatic events (McGibbon et al., 2018). We suggest that a deep-seated source for lixiviants such as CO_2 that leach U can explain the low $\delta^{238}U$ values in the travertine calcite. Figures S1 and S2 in the Supplemental Material each show a vein that represents a paleo-path of upwelling CO_2-rich travertine-depositing groundwater. These figures also show the veins' $\delta^{238}U$ time series and age. Figure S3 offers profiles of the regional general geology associated with these upwelling spring waters. All data are in Tables S1 and S2.

DISCUSSION

The −2.85‰ to −0.83‰ $\delta^{238}U$ value of spring water forming travertine at Soda Dam and Tierra Amarilla anticline shows that the uranium anomaly is carried by the water and not produced at the surface during travertine deposition. Also, none of our sites showing this anomaly appear to be influenced by large bodies of aquifer water, such that these spring systems are issuing water that has not been greatly diluted. In this study, leaching is proposed as the mechanism producing the exceedingly low $\delta^{238}U$ values in a simple two-component system where magmatic-driven carbonic water is rising toward the surface from depth having low U, low $\delta^{238}U$, low $\delta^{234}U$, and igneous $^{87}Sr/^{86}Sr$, and near the surface it mixes with meteoric groundwater in Precambrian granite, Paleozoic limestones, and Mesozoic sandstones. U concentration, $\delta^{234}U$, $\delta^{238}U$, $^{87}Sr/^{86}Sr$, and alkalinity of water samples along the Nacimiento Fault show weak correlations (Fig. 3) that agree with these primal upwelling waters being only lightly diluted with groundwater having lower CO_2, higher U concentration, bulk-earth $\delta^{238}U$ values, high $\delta^{234}U$, and high granitic $^{87}Sr/^{86}Sr$ values. Most of the mixing probably takes place in the thicker Precambrian basement rocks, consistent with our most robust correlation in our dataset between $\delta^{238}U$ and $^{87}Sr/^{86}Sr$ (Fig. 3D). We favor an interpretation that upwelling dissolved CO_2 is leaching ^{235}U preferentially over ^{238}U in deep-seated carbonic spring systems.

Routes of these spring waters via the western Rio Grande rift and Valles Caldera joints and faults are envisioned to be deep seated and associated with abundant magmatic- and/or mantle-sourced CO_2 (Crossey et al., 2009; Gilfillan et al., 2008). Deep-seated endogenic systems involve anoxic fluids along flow paths that deliver reduced chemical species and microbes (Crossey et al., 2016) to the more oxidized environments at the surface (Des Marais and Walter, 2019). Neither adsorption and reduction-oxidation occurring at the surface just prior to nor during calcite precipitation, as evidenced by ferromanganese materials and Fe-rich fossil bacteria in the Fe-oxide-rich travertine deposits and overall lower $\delta^{234}U$ values for the travertine versus the spring water explain the similarly low $\delta^{238}U$ values in both the water and travertine. The $\delta^{238}U$ anomaly is clearly produced in the subsurface. The interpretation we favor explaining the low $\delta^{238}U$ values in the spring water and travertine involves preferential stripping of the lighter ^{235}U isotope along a long travel path in a reduced environment by CO_2-rich fluids.

CO_2 is an effective U lixiviant (leach solution), and CO_2 lixiviation is a process by which U is or has been extracted in in-situ mining (Tweeton and Peterson, 1981). A favored process for stripping, oxidizing, and transporting U in in-situ mining utilizes supercritical CO_2 mixed with tri-*n*-butylphosphate (Sawada et al., 2006). CO_2-rich magma- and/or mantle-derived volatiles are common in springs in the Rio Grande rift and on the Colorado Plateau (Crossey et al., 2009, 2016; Newell et al., 2005). CO_2 lixiviation is the process we favor for sufficient leaching of U from various rock types over the long CO_2 travel path in a reduced joint or fracture environment. The stripping of U by CO_2 in a deep setting probably offsets the lower solubility of U in reduced conditions (Langmuir, 1997) of that deep setting. Bigeleisen (1996) showed that ^{235}U behaves in a chemical exchange in a reduced environment as though it has a smaller atomic mass, leaving ^{238}U more concentrated in the U(IV) material species. In this scenario, the reduced environment preferentially favors beyond-normal mass-dependent leaching of ^{235}U over ^{238}U, resulting in transport of water having exceptionally negative $\delta^{238}U$ values to the surface and deposition of travertine that inherits those low values. These low $\delta^{238}U$ values of travertine along the western margin of the Rio Grande rift and of extinct travertine systems are therefore potential indicators of magma- and/or mantle-sourced CO_2 systems that were active in the past as well. The lesser anomalies measured in the travertine at the Belen and Mesa del Oro sites may represent a system that has mixed with greater amounts of meteoric water at moderately shallow depths and/or less CO_2.

Terrestrial $\delta^{238}U$ values comparable to those of these spring waters and travertine calcite are from groundwaters downstream of Australian sandstone-hosted U deposits and were due to reduction-oxidation and U mineralization in a shallow system (Murphy et al., 2014). Our exceptionally low $\delta^{238}U$ values (−3.2‰ to −0.8‰) in water sourced from depth and in travertine deposits are interpreted as products of leaching that involve natural lixiviants such as CO_2 generated by magmatic activity rather than surface or near-surface reduction-oxidation or travertine precipitation (degassing) processes. $\delta^{238}U$ values of similar endogenic-type travertine deposits globally should share comparable

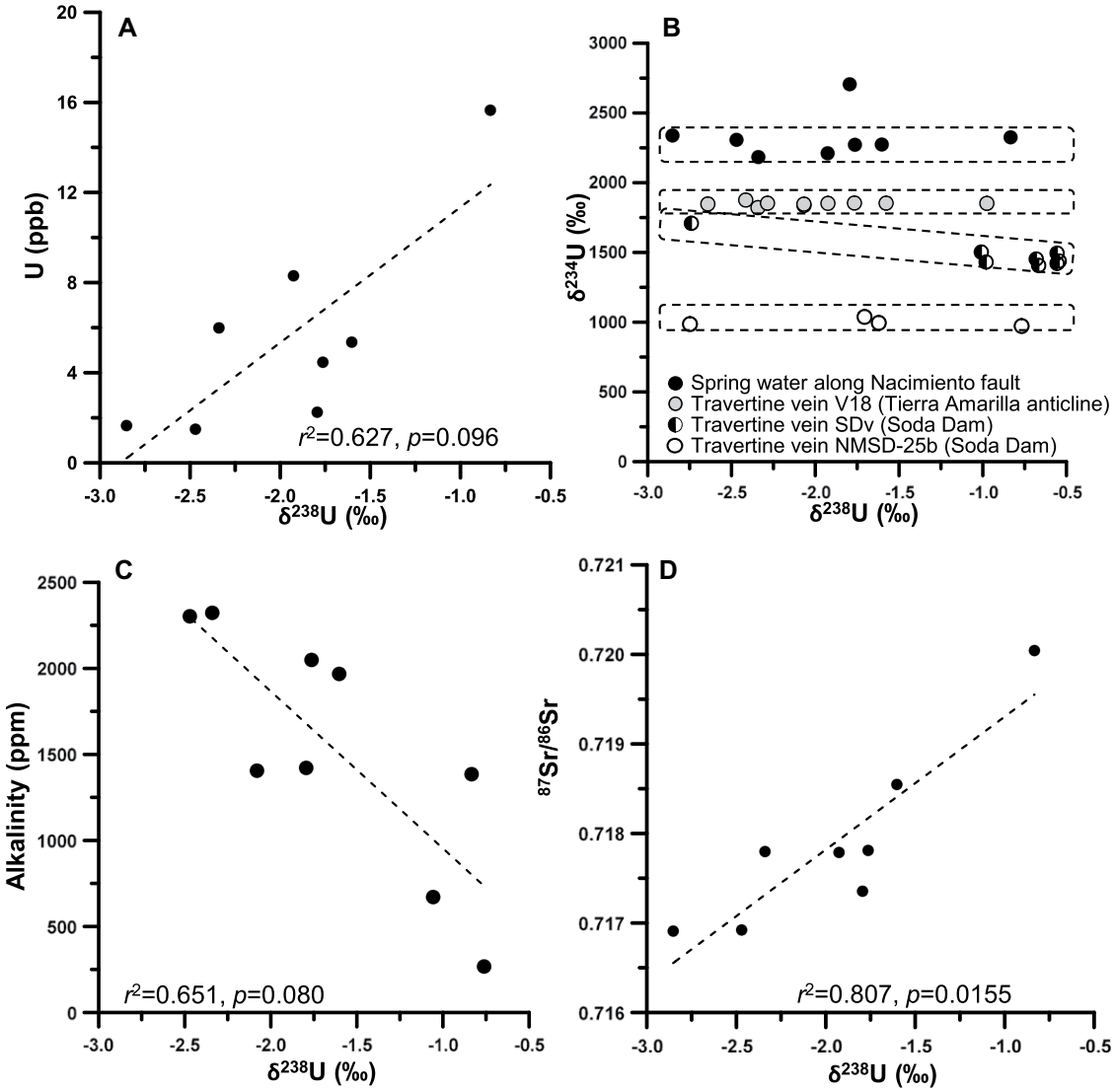

Figure 3. U and Sr elemental and isotopic values for spring water and travertine. (A) $\delta^{238}U$ versus U concentration of spring water along Nacimiento fault shows weak positive correlation, which has been previously identified as a proxy for redox conditions (Bopp et al., 2010). (B) $\delta^{238}U$ versus $\delta^{234}U$ of spring water along Nacimiento fault and of three travertine veins having multiple analyses. Results seem to show no significant change in $\delta^{234}U$ relative to $\delta^{238}U$, as indicated by the dashed boxes. (C) $\delta^{238}U$ versus alkalinity shows less-than-robust negative correlation for spring waters along Nacimiento fault. (D) Same spring waters as in C show robust correlation between $\delta^{238}U$ and $^{87}Sr/^{86}Sr$.

U-isotope signatures, and fossil travertines with these values could yield evidence of upwelling CO_2 systems associated with magmatic or tectonic activity in deep time of tens to hundreds of millions of years.

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