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Authors

Morrissey, Sheila K
Clark, Jordan F
Bennett, Michael
et al.

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Groundwater reorganization in the Floridan aquifer following Holocene sea-level rise

Sheila K. Morrissey¹, Jordan F. Clark^{1*}, Michael Bennett², Emily Richardson³ and Martin Stute⁴

Sea-level fluctuations, particularly those associated with glacial-interglacial cycles, can have profound impacts on the flow and circulation of coastal groundwater: the water found at present in many coastal aquifers may have been recharged during the last glacial period, when sea level was over 100 m lower than present^{1,2}, and thus is not in equilibrium with present recharge conditions³⁻⁹. Here we show that the geochemistry of the groundwater found in the Floridan Aquifer System in south Florida is best explained by a reorganization of groundwater flow following the sea-level rise at the end of the Last Glacial Maximum approximately 18,000 years ago. We find that the geochemistry of the fresh water found in the upper aquifers at present is consistent with recharge from meteoric water during the last glacial period. The lower aquifer, however, consists of post-sea-level-rise salt water that is most similar to that of the Straits of Florida, though with some dilution from the residual fresh water from the last glacial period circulation. We therefore suggest that during the last glacial period, the entire Floridan Aquifer System was recharged with meteoric waters. After sea level rose, the increased hydraulic head reduced the velocity of the groundwater flow. This velocity reduction trapped the fresh water in the upper aquifers and initiated saltwater circulation in the lower aquifer.

Groundwater recharged during the last glacial period (LGP), which is geochemically different from groundwater recharged during the Holocene, still exists today in many aquifers and can be used to determine how climate change affected groundwater flow. Geochemical data (see Supplementary Information), including total dissolved solids (TDS), stable isotopes of water ($\delta^{18}\text{O}$ and δD) and dissolved noble gases, were analysed using standard methods to evaluate groundwater flow dynamics and to establish residence times in the three coastal aquifers of the confined Floridan Aquifer System (FAS) in south Florida: the Upper Floridan Aquifer (UFA), the Avon Park Permeable Zone (APPZ) and the Lower Floridan Aquifer (LFA), which together comprise about 800 m of permeable Tertiary carbonate (limestone and dolostone) sediments^{8,9}. The aquifer system is relatively shallow in the north, where it is hydraulically connected with the surface, and deepens to the south of the study area (Fig. 1). Separating the upper and lower aquifers are semi-permeable layers of high-magnesium limestone and dolostone, which in some areas may transmit water vertically between the two aquifers depending on the difference in hydraulic head⁸⁻¹⁰.

The groundwater that exists today in the FAS can be divided into four groups on the basis of salinity and relative age (Table 1, Fig. 1) and into two major flow systems. The UFA and APPZ are recharged by precipitation in central Florida north of Lake Okeechobee, where the aquifers are hydraulically connected to an unconfined surficial

aquifer⁸. In the UFA and APPZ, which are dominated by fresh water, groundwater flows south from the recharge area towards both coasts, and shows increasing TDS with distance along its flowpath. Fresh water also exists in the LFA near the recharge area. However, the remainder of the LFA groundwater is saline, and much of it is comparable in salinity to the sea water in the nearby Straits of Florida.

Recharge temperatures, excess air and fractionation factor were calculated from dissolved noble-gas data with the NOBLEGAS computer model using the closed equilibrium model and end-member salinities^{11,12}. Excess air concentrations as represented by ΔNe concentrations were typical (Table 1) and did not correlate with recharge temperature and therefore age. Previously published records determined from noble-gas palaeo-thermometry indicate an approximate 5 °C temperature difference between LGP- and Holocene-recharged groundwaters in the southeastern United States, with noble-gas recharge temperatures (NGRTs) within about 2 °C of the mean annual air temperature (MAAT) in the recharge area¹³⁻¹⁶. Following the method of Clark *et al.*¹⁵, the lower NGRT of FAS groundwater was used as a 'stratigraphic' marker to identify glacial-aged groundwater in the freshwater flow system. This was later verified by examining relative ages determined with helium (He) concentrations, which increase with time as a result of uranium and thorium series decay within the aquifer and deeper within the Earth's crust¹⁷⁻¹⁹, thereby making excess He (the portion of He concentration greater than the solubility equilibrium) a useful chronometer^{13-16,20}.

In the UFA and APPZ, recharge-area wells were found to have NGRTs (mean = 24.7 ± 1.2 °C) similar to the local MAAT (22.5 °C; mean of ten stations in the recharge area from www.sercc.com). NGRTs calculated from groundwater collected from the surficial aquifers (22.7–26.7 °C; $n = 2$) are warmer than MAAT, consistent with Holocene-aged recharge. Groundwater from most of the remaining UFA and APPZ wells has NGRTs that are about 5 °C cooler than the Holocene-recharged water (mean = 19.7 ± 1.1 °C), reflecting the cooler climate of the LGP (Fig. 2). In the LFA, recharge-area fresh water was calculated to have a Holocene-like NGRT (mean = 25.5 ± 1.0 °C) whereas more southern groundwater has salinities greater than 30,000 ppm and recharge temperatures (mean = 9.1 ± 0.8 °C; omitting well OSF-104L) similar to the water temperature at the bottom of the Straits of Florida (7.8–12.2 °C). Recent measurements of sea water have shown that, for intermediate waters, noble-gas concentrations are within 2% of the equilibrium concentration at the *in situ* temperature²¹. One freshwater LFA well immediately south of the recharge area (Romp 28-4; 22.0 °C) was intermediate between the two endmembers, most likely resulting from mixing of different age waters within its 30-m-long screen interval.

¹Department of Earth Science, University of California, Santa Barbara, California 93106, USA, ²AECOM 2990 Palm Beach Lakes Boulevard, West Palm Beach, Florida 33409, USA, ³South Florida Water Management District, 3301 Gun Club Road, West Palm Beach, Florida 33406, USA, ⁴Lamont-Doherty Earth Observatory, Columbia University, Palisades, New York 10964, USA. *e-mail: jfclark@geol.ucsb.edu.

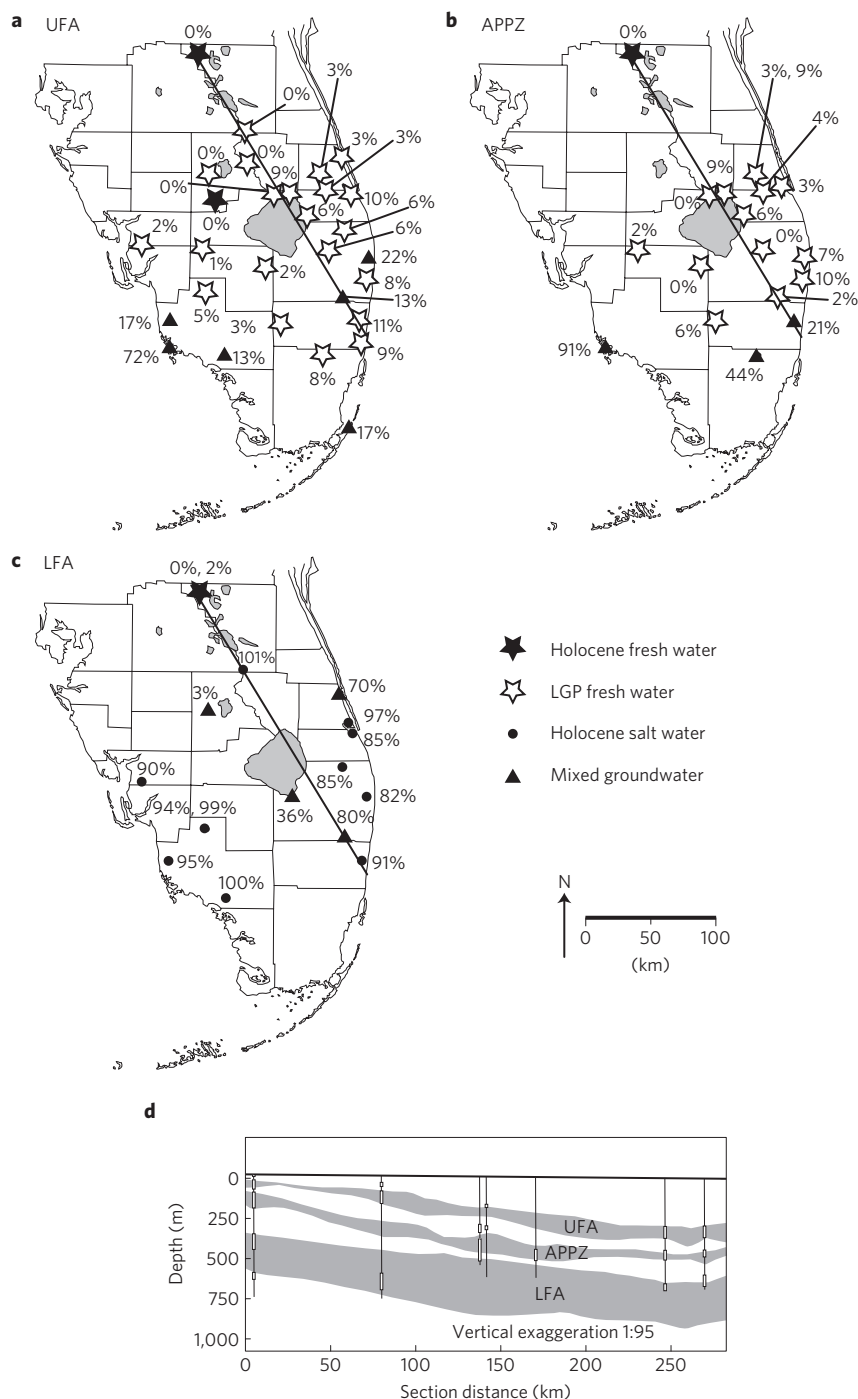


Figure 1 | Maps of wells and cross-section through the field area. a–c, Maps of south Florida with sampling locations in the three aquifers with each well marked on the basis of its geochemical group. Mixed groundwater has been inferred from either salinity or noble-gas recharge temperature. Seawater mixing percentages were calculated using 1,000 ppm and 34,940 ppm for the freshwater and seawater endmembers; well water fresher than 1,000 ppm was assigned 0%. At the multi-level wells, the shallower intervals are listed first. The solid line extending from the top of the maps to the southeast represents the location of the cross-section shown in **d**. **d,** Aquifer cross-section. The vertical lines (open area = screened zones) represent the location of wells within 9 km of the section.

During the LGP, the $\delta^{18}\text{O}$ of the ocean water was enriched by approximately 1‰ compared with the modern ocean because lighter isotopes were preferentially transported to ice sheets^{22,23}. This change was transferred through the water cycle causing a 1‰ enrichment in precipitation sourced from the LGP ocean. Although the data ranges overlap, the LGP-recharged fresh water (mean = $-1.7 \pm 0.3\text{‰}$ $\delta^{18}\text{O}$) is, on average, enriched compared with the Holocene-recharged fresh water (mean = $-2.3 \pm 0.7\text{‰}$ $\delta^{18}\text{O}$)

by an amount similar to the known enrichment of the LGP ocean (Fig. 3). Similar enrichments in LGP-recharged groundwater have been reported in a coastal aquifer in Portugal²⁰ and in the FAS north of the study area⁷.

The LFA salt water has $\delta^{18}\text{O}$ values (mean = $+0.2 \pm 0.1\text{‰}$) similar to the $\delta^{18}\text{O}$ of recently collected Straits of Florida bottom water ($+0.3$ to $+0.5\text{‰}$ below 450 m; Table 1; Fig. 3). Foraminifera tests from sediments collected near the base of the Florida platform

Table 1 | Ranges for geochemical parameters in each of four groundwater groups and Straits of Florida.

	Holocene fresh water	LGP fresh water	Holocene salt water	Mixed groundwater*	Straits of Florida
TDS (ppm)	200–4,500	270–4,750	29,000–36,200	5,400–26,100	34,940
Noble-gas recharge T ($^{\circ}\text{C}$)	23.2–26.2	18.0–21.8	8.0–10.2 [†]	–	7.8–12.2 [‡]
$\delta^{18}\text{O}$ (‰)	–3.4 to –1.5	–2.8 to –1.0	+0.0 to +0.4	–1.5 to –0.1	+0.3
ΔNe (‰)	8–649	34–428	9–55	–	–
Excess He (10^{-8} cm^3 STP g^{-1})	0.4–6.3	3.1–36	3.1 to 13.3	9.2–33	–
^{14}C (pmc)	4.3–10.5	0.5–42.6	0.3–19.5	0.6–1.8	–

*NGRTs and ΔNe are not reported for the mixed groundwater samples.

[†]This excludes the NGRT in OFS-104L, which was 13.2 $^{\circ}\text{C}$.

[‡]Measured water temperature for water depths below 450 m at 27 $^{\circ}$ 0.445 N 79 $^{\circ}$ 36.789 W.

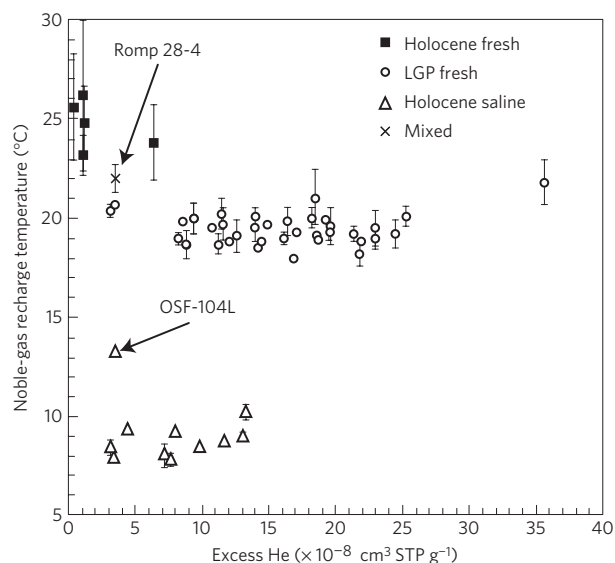


Figure 2 | NGRTs and relative ages expressed as a function of excess helium concentrations. The error bars denote the uncertainty of the noble-gas fit calculated with the NOBLEGAS model; in many cases the error bar is smaller than the symbol. See Supplementary Fig. S1 for the location of OSF-104 and Romp 28-4.

show an enrichment in $\delta^{18}\text{O}$ by about 1.2‰ during the Last Glacial Maximum compared with the Holocene²⁴, reflecting both the transfer of $\delta^{18}\text{O}$ -depleted water into ice sheets and cooler ocean water temperatures. If the salt water in the LFA infiltrated during the LGP, it should be isotopically enriched and have cooler NGRTs than the bottom water found at present in the Straits of Florida. Instead, the similar geochemistry of the saline groundwater and modern ocean water indicates that Holocene-aged sea water is circulating through the LFA. It is important to note that much of the groundwater in the LFA is fresher and slightly more depleted in $\delta^{18}\text{O}$ than the ocean water source. On the basis of the $\delta^{18}\text{O}$ and TDS values, the likely freshwater source is LGP groundwater (Fig. 3).

Excess He concentrations in FAS groundwater samples were used to determine relative ages. No numerical ages were assigned because of uncertainties associated with the accumulation rate of He in the groundwater. In the UFA and APPZ, excess He concentrations are lowest (youngest ages) in wells below and just down gradient of the recharge area, whereas higher excess He concentrations (older ages) were found in wells further down gradient. LFA excess He concentrations are also low in the recharge area. These distributions reflect the known circulation patterns in the FAS. At all locations with multi-level wells, He excesses in the saline LFA samples were less than the excesses in the overlying fresh water sampled from the UFA and APPZ. The relative chronology

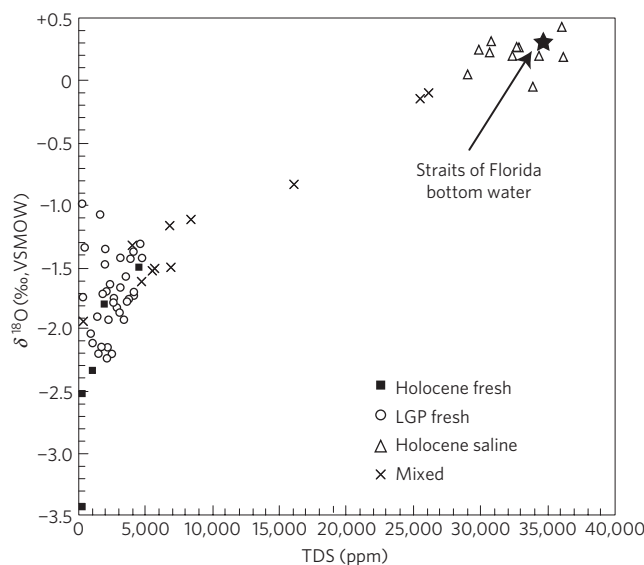


Figure 3 | $\delta^{18}\text{O}$ as a function of salinity (as measured by TDS). Straits of Florida bottom water is denoted as a star and is similar to the Holocene saltwater samples. The mixed groundwater samples have $\delta^{18}\text{O}$ values that fall along a mixing line between Straits of Florida bottom and the LGP fresh water.

based on excess He agrees well with the age groups determined with noble-gas recharge temperatures; Holocene-aged groundwater has lower excess He concentrations (Fig. 2).

Kohout²⁵ hypothesized that the LFA south of Lake Okeechobee is recharged by coastal sea water entering the aquifer from near the bottom of the Straits of Florida at a depth of about 600 m. This water flows inland beneath the discharging UFA fresh water in the highly permeable section of the LFA referred to as the Boulder Zone. The density difference between sea water and discharging fresh water alone would be sufficient to cause cyclic flow of groundwater in coastal aquifers²⁶. However, the Kohout flow hypothesis was based on temperature data indicating that the salt water is geothermally heated as it moves inland through the LFA; and the consequent decrease in fluid density causes the salt water to rise in the LFA, where it leaks into the upper aquifers and is transported to the ocean by the freshwater flow system²⁷. At present, the flux of salt water into the LFA is controlled by both the permeability²⁶ and the geometry²⁸ of the carbonate Florida platform. It has been hypothesized that the flux of sea water into the LFA and its circulation are also influenced by changes in sea level and consequent changes in groundwater gradient^{7,8}.

The geochemical data provide a documentation of groundwater circulation changes in the FAS of south Florida resulting from sea-level rise. At the Last Glacial Maximum, the global sea level

was approximately 120 m lower than today^{1,2} and the land area of the shallow Florida platform was nearly doubled. Nearly all of the land-area gain was due to a shift of Florida's west coast more than 100 km into the Gulf of Mexico. The lower sea level during the LGP undoubtedly caused increased hydraulic gradients of groundwater in the FAS, resulting in faster groundwater flow rates compared with today's rates in both the UFA and APPZ, assuming that the water table was not significantly lower during the LGP. If it were lower, then the NGRTs would be higher owing to geothermal heating of the soil and the temperature difference less than 5 °C. We hypothesize that fresh water was circulating through the LFA during the LGP. This is supported by the small component of LGP fresh water that exists there today and mixes with the circulating salt water causing a slight decrease in the salinity and $\delta^{18}\text{O}$ of the LFA groundwater relative to the ocean. It is unlikely that this LGP fresh water recently leaked down from the overlying freshwater-filled aquifers given the known upward flow of salt water from the LFA.

Approximately 18,000 years ago, the climate warmed and sea level rose^{1,2}, flooding the Gulf of Mexico side of the Florida platform and increasing the hydraulic head of sea water along Florida's coasts. The resulting >80% decrease in groundwater gradient slowed the flow of fresh water suddenly and significantly, trapping it in the UFA and APPZ. Today the UFA and APPZ primarily contain this slowly moving LGP fresh water with its relatively uniform $\delta^{18}\text{O}$ values near -1.7‰ and NGRTs near 19.4 °C (Table 1). Similar geochemical evidence for the trapping of LGP groundwater in coastal aquifers has been reported elsewhere^{3,7,15,20}. Close to the recharge area the LGP fresh water transitions abruptly to Holocene fresh water with $\delta^{18}\text{O}$ values near -2.3‰ and NGRTs near 24.7 °C. The increased seawater head also forced sea water into the LFA at the Boulder Zone, flushing LGP fresh water from the aquifer and initiating saltwater circulation. Most of the LFA is now filled with Holocene-recharged salt water with geochemical properties similar to the sea water in the Straits of Florida (Table 1). The associated changes to groundwater flow resulting from glacial sea level rise, which are documented here with geochemical tracers, are especially evident in south Florida, where the steep coastline along the south coast of the Florida platform and low inland topography exaggerated the hydrologic effects of sea-level change.

We document that most of the fresh water in the upper Floridan aquifers in south Florida (south of Lake Okeechobee) recharged during the LGP. This water, which is below the major population centres, is targeted as a new potable source. Our results call the long-term sustainability of this source into question.

Methods

Groundwater samples were collected from wells in the surficial aquifers ($n = 11$), UFA ($n = 32$), APPZ ($n = 21$) and LFA ($n = 16$), across south Florida between October 2000 and December 2007. Four wells were sampled twice. In addition, one sample was collected from a surface water canal. At least three well volumes were purged from each well before sample collection. All wells were pumped using a suction pump, except for artesian wells, which were allowed to flow freely.

All samples were tested in the field for temperature, conductivity and pH. In addition, water collected from each well was analysed for inorganic ions (Na^+ , K^+ , Ca^{2+} , Mg^{2+} , Cl^- , SO_4^{2-} and SiO_2), stable isotopes of water ($^{18}\text{O}/^{16}\text{O}$ and D/H), dissolved noble gases (He, Ne, Ar, Kr and Xe) and dissolved inorganic carbon isotopes ($^{13}\text{C}/^{12}\text{C}$ and ^{14}C). See the Supplementary Information for a brief discussion of the ^{14}C data. Samples collected in 2007 were also analysed for tritium (T).

Inorganic ion samples were collected in 250 ml plastic bottles and analysed by the South Florida Water Management District's contract laboratories. Samples analysed for stable isotopes of water were collected in 60 ml glass bottles and sent to the University of California Davis for analysis using standard mass spectrometry. The results are presented in δ -notation relative to the Vienna Standard Mean Ocean Water (VSMOW)

$$\delta^{18}\text{O} \text{ or } \delta\text{D} = \left(\frac{R}{R_{\text{VSMOW}}} - 1 \right) \times 1,000$$

where R is the isotope ratio of the sample and R_{VSMOW} is the isotope ratio of VSMOW. The $\delta^{18}\text{O}$ and δD results have errors of $\pm 0.05\text{‰}$ and $\pm 0.5\text{‰}$, respectively.

Dissolved noble-gas samples were collected in 20 ml copper tubes that were connected to the pump outlet by a short piece of Tygon tubing. A clear plastic tube with a needle valve was attached to the outlet end of the copper tube during sampling. The copper tubes were flushed at high pressure to eliminate bubbles by reducing the flow through the valve until no bubbles were observed. The copper tubes were sealed with stainless-steel pinch-off clamps and sent to the Lamont-Doherty Earth Observatory at Columbia University for analysis on a MAP 215-50 noble-gas mass spectrometer using the methods of ref. 14. Absolute concentrations of the noble gases were determined with errors of $\pm 1\%$ for Ar and Xe, and $\pm 2\%$ for He, Ne and Kr.

Dissolved inorganic carbon samples were collected in 250 ml glass bottles. At the University of California Santa Barbara, carbon was extracted from each sample on a vacuum line by adding approximately 15 ml of degassed 1 N hydrochloric acid (HCl) and vacuum stripping the water. The liberated CO_2 was trapped, separated from water vapour and split for $^{13}\text{C}/^{12}\text{C}$ and ^{14}C analyses. $^{13}\text{C}/^{12}\text{C}$ ratios were determined using a conventional mass spectrometer and reported in δ -notation relative to the Vienna Pee Dee Belemnite (VPDB) standard:

$$\delta^{13}\text{C} = \left(\frac{R}{R_{\text{VPDB}}} - 1 \right) \times 1,000$$

where R is the isotope ratio of the sample and R_{VPDB} is the isotope ratio of VPDB. The $\delta^{13}\text{C}$ errors are $\pm 0.3\text{‰}$. The radiocarbon contents were determined by accelerated mass spectrometry after the CO_2 gas had been reduced to graphite at University of Arizona (2000 and 2001 samples) and at University of California Irvine (2005 and 2007 samples). The radiocarbon abundances were reported as per cent modern carbon (pmc) and have errors of ± 1 pmc.

Samples analysed for tritium were collected in 500 ml plastic bottles and sent to University of Waterloo Environmental Isotope Laboratory for analysis. Following isotope enrichment, the tritium isotope concentrations were measured by liquid scintillation. The tritium concentrations were reported in tritium units (TU), where one TU equals 1 T atom per 10^{18} hydrogen atoms, and have errors of ± 0.5 TU and a detection limit of 0.8 TU.

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Author contributions

J.F.C. and M.B. conceived the study. M.B. and E.R. organized the field collection events. M.S. conducted the noble-gas analyses. J.F.C., S.K.M., M.B. and E.R. implemented the other geochemical analyses. All authors participated in data discussion and interpretation. S.K.M. and J.F.C. wrote the initial manuscript, and all authors provided substantial comments and editorial revisions to the manuscript.

Additional information

The authors declare no competing financial interests. Supplementary information accompanies this paper on www.nature.com/naturegeoscience. Reprints and permissions information is available online at <http://npg.nature.com/reprintsandpermissions>. Correspondence and requests for materials should be addressed to J.F.C.