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Supplementary Materials for

Strong links between Saharan dust fluxes, monsoon strength, and North Atlantic climate during the last 5000 years

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The PDF file includes:

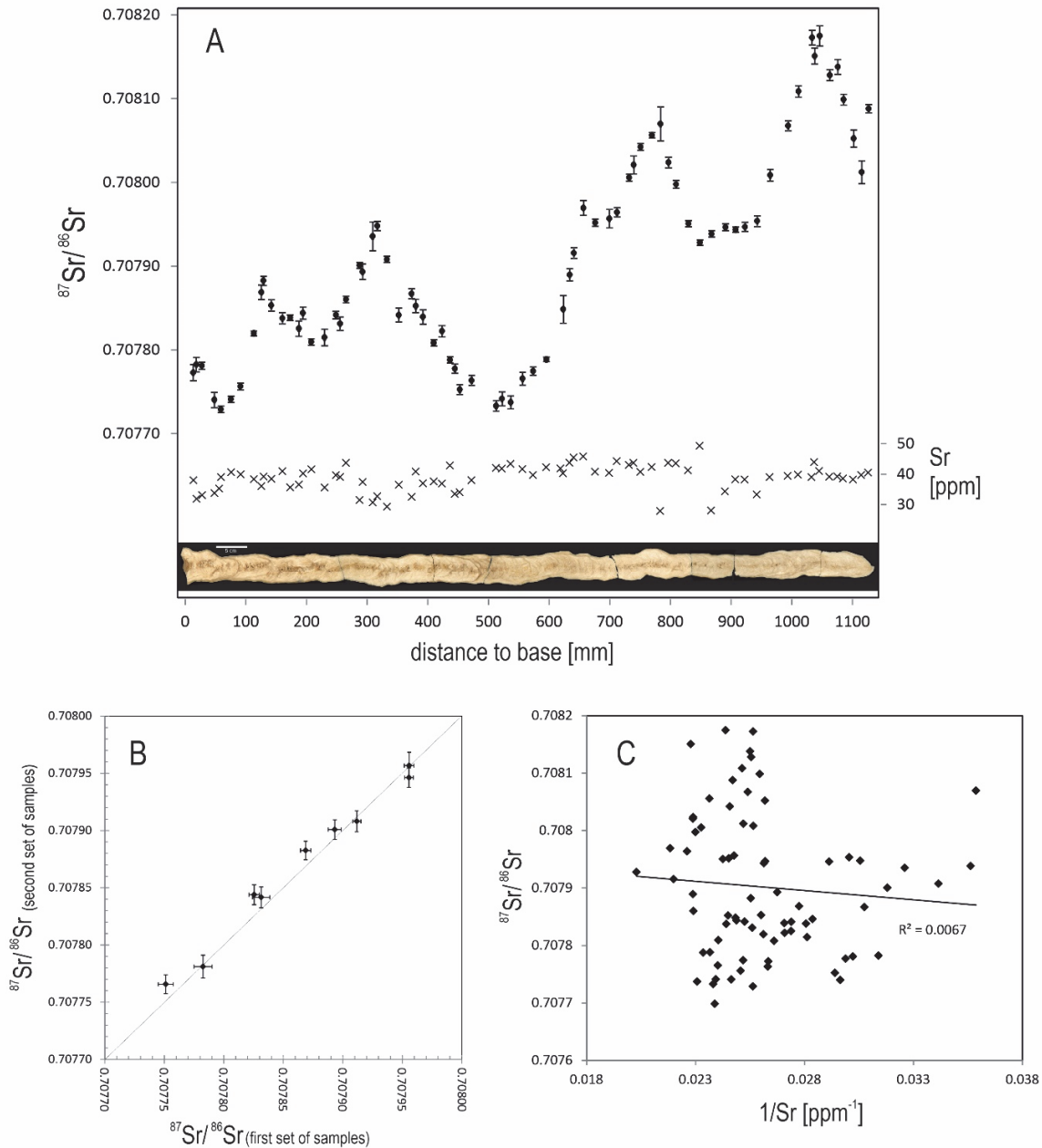
Figs. S1 to S4
Tables S1 and S2
Legend for data file S1
References

Other Supplementary Material for this manuscript includes the following:

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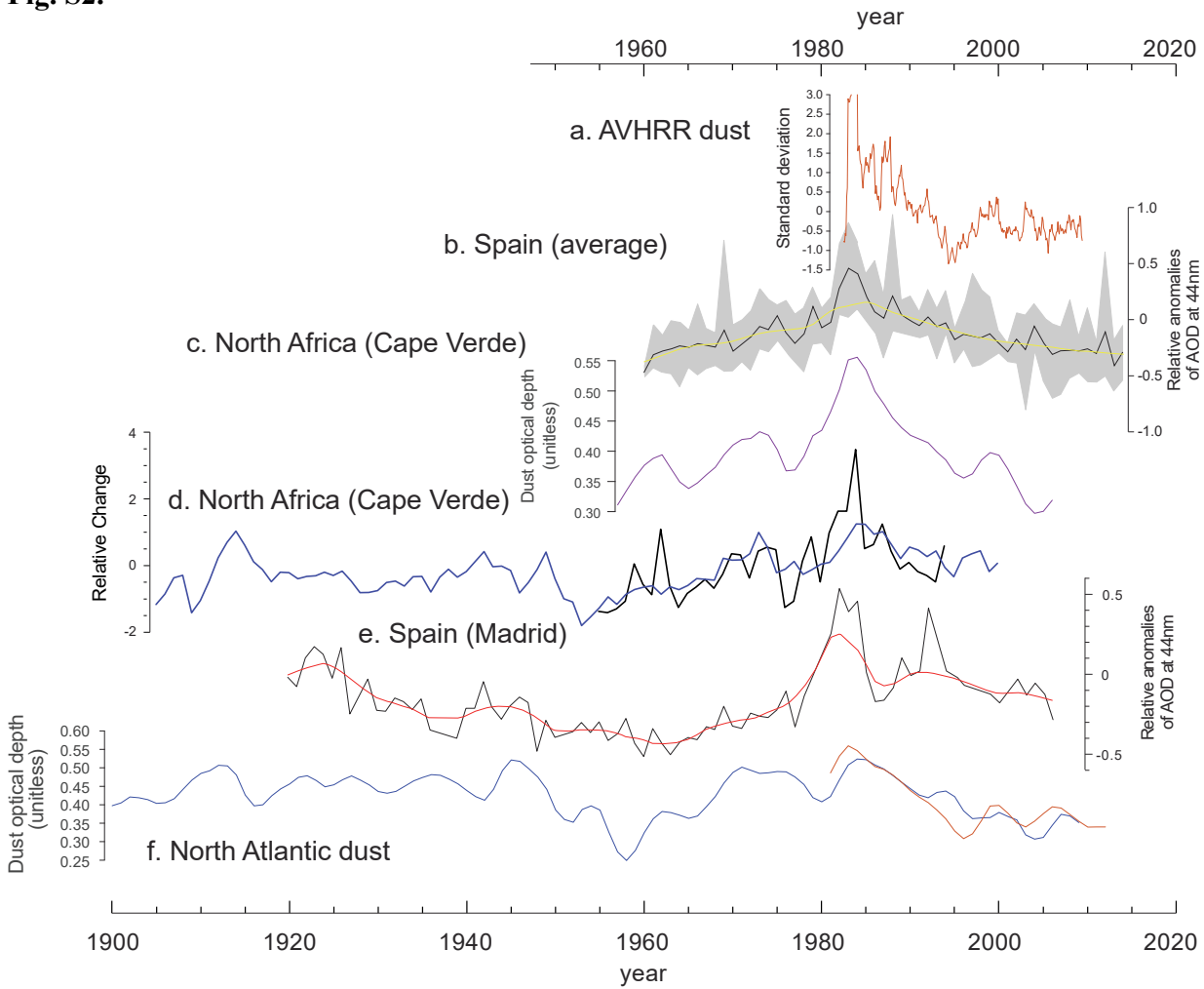
Data file S1

Fig. S1.



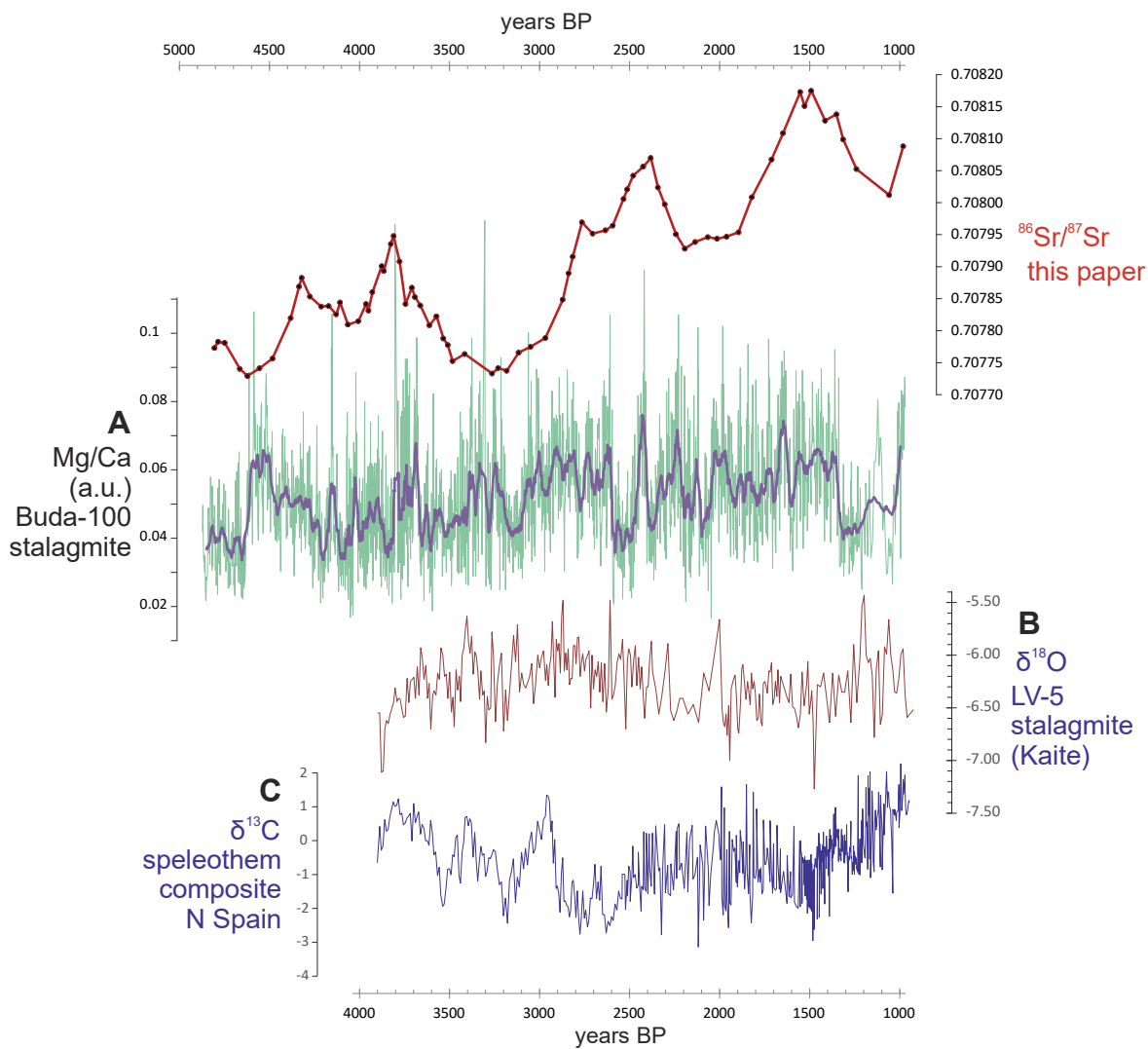
$^{87}\text{Sr}/^{86}\text{Sr}$ analytical results in Buda-100 stalagmite. **A)** $^{87}\text{Sr}/^{86}\text{Sr}$ ratios vs. stratigraphic position, represented by the vertical distance to the base of the stalagmite. The plot also includes the bulk strontium concentration of the same samples used for isotope measurements, which does not any clear trend nor variability pattern. **B)** Replicability of $^{87}\text{Sr}/^{86}\text{Sr}$ measurements checked by comparison of results of nine pairs of samples extracted from approximately the same stratigraphic levels. Linear correlation indicates excellent replicability. Observed minor deviations are related to the fact that each pair does not consists of two aliquots of one sample but to independently extracted samples. **C)** $^{87}\text{Sr}/^{86}\text{Sr}$ ratios vs. strontium concentration ($1/\text{Sr}$) showing lack of significant correlation between the two variables.

Fig. S2.



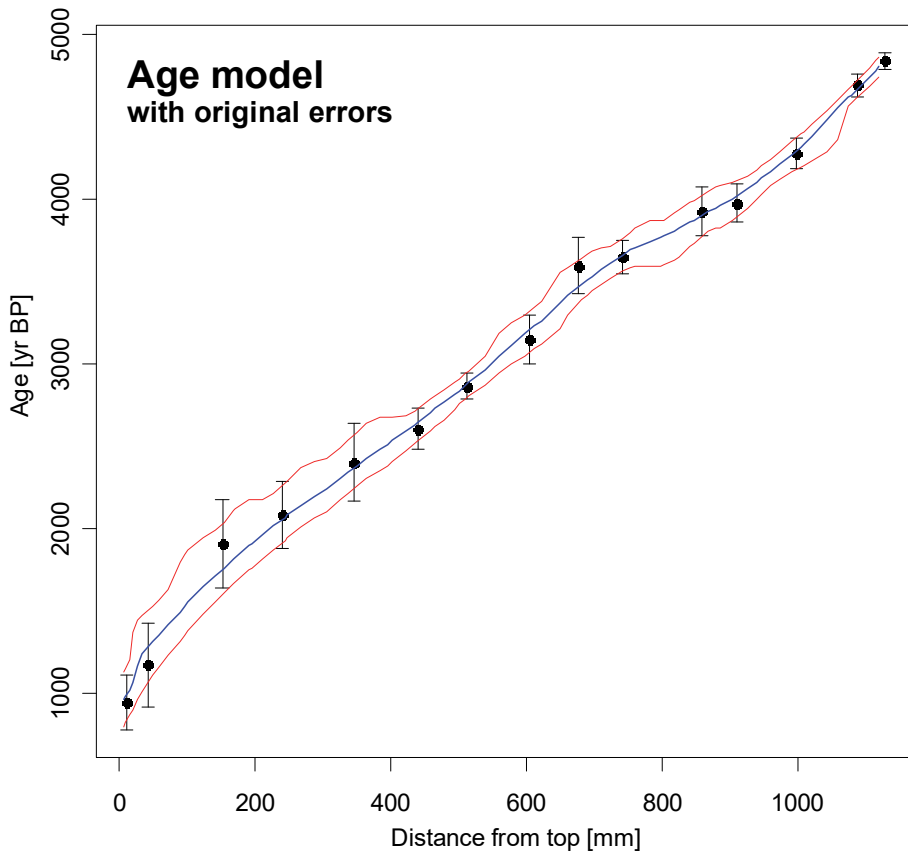
Inferred changes in dust influxes over Spain compared to NW Africa during the instrumental period. a) Dust optical depth averaged over the tropical North Atlantic from Advanced Very High Resolution Radiometer space-borne (AVHRR dust) (81), **b)** Mean relative anomalies of estimated AOD₄₄₀ for summer (black line) for eight sites in Spain for 1961–2014 interval (yellow line: 11 years moving average; shaded area: maximum range for each year) (82), **c)** Hybrid satellite – coral reconstruction over the Cape Verde islands (15° N, 23.5° W) (83), **d)** Estimated relative dustiness for North Africa and North Atlantic. Cape Verde relative dustiness is shown in black, while North African dustiness (extrapolated using the Palmer Drought Severity Index-PDSI) is shown in blue (84); **e)** Relative anomalies of estimated AOD at 440 nm (black line) for Madrid (central Spain) in summer for 1920–2007 period (red line: 11-years moving average) (82). **f)** Estimates of North Atlantic dust (PC2 time series from the CIRES-20CR and ERA-I reanalyses) (81).

Fig. S3.



Paleoclimate records from Kaite Cave for the 5.0 to 1.0 ka BP interval, compared with the $^{86}\text{Sr}/^{87}\text{Sr}$ record. **A)** Mg/Ca series from stalagmite Buda-100, proxy of local hydroclimate, with higher values indicating drier conditions (45). Relative changes are however interpreted to be low. **B)** Calcite $\delta^{18}\text{O}$ of Kaite speleothem LV5 (85), with age model from (86). Changes in $\delta^{18}\text{O}$ have been interpreted to result from changes in precipitation amount, with higher values indicating lower amount (87), or alternatively from changes in seasonal rainfall patterns, with higher values indicating higher summer vs. winter rainfall (44). **C)** Calcite $\delta^{13}\text{C}$ composite curve from three speleothems from N Spain caves (including Kaite Cave) considered to reflect changes in surface temperature (86). The lack of correlation of these three records with the $^{86}\text{Sr}/^{87}\text{Sr}$ series, points in the same direction than the available instrumental series of dust influxes over Spain (Fig. S1) and the supra-regional correlations (Fig. 4): the dust changes detected in stalagmite Buda-100 are dominantly reflecting broad changes in dust source areas, being the influence of possible changes in regional dust transport or deposition subordinated.

Fig. S4.



Age model for stalagmite Buda 100. Based in data from Table S2 and performed with StalAge software (68).

Table S1.

		$^{87}\text{Sr}/^{86}\text{Sr}$	Avg. Error	Number*	Std Dev
Present day speleothems	high growth	0.708386	$\pm 2.1\text{E-}06$	4	1.7E-06
	low growth	0.708295	$\pm 2.0\text{E-}06$	3	3.0E-05
Stalagmite	buda 100	0.707912	$\pm 6.8\text{E-}06$	89	1.2E-04
Drip waters	fast drip	0.708378	$\pm 2.5\text{E-}06$	4	3.7E-06
	slow drip	0.708301	$\pm 2.8\text{E-}06$	4	1.2E-05
Lithosol	soil leachate	0.708203	$\pm 6.2\text{E-}06$	4	1.5E-05
Host rock	host rock	0.707386	$\pm 5.2\text{E-}06$	8	3.3E-05
Sea spray	sea salt	0.709175	-	**	**
Rain	<i>clean</i> rain	0.709086	$\pm 2.4\text{E-}06$	4	5.8E-04
	<i>dusty</i> rain	0.720912	$\pm 3.3\text{E-}06$	5	1.3E-03
Saharan dust	Western North African PSA	0.7279	-	***	5.2E-03
	Central North African PSA	0.7186	-	***	5.3E-03
	Canary Islands accum.	0.725225	-	****	****

* Number of analyzed samples

**Sea salt isotope ratio according to (70).

***Isotope ratio for Western and Central North African preferential source dust areas (PSAs) according to (20)

****Isotope ratio for Canary Islands Saharan dust accumulations based in (18, 71).

$^{87}\text{Sr}/^{86}\text{Sr}$ in Kaite Cave and speleothem strontium sources. The table includes averaged data for the sources of strontium (host rock, sea spray, Saharan dust) determining the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the speleothems and included in the model for dust influx reconstruction. It also includes the isotopic ratios of present-day drip-waters and growing speleothems, necessary for model calibration, and the mean isotopic ratios measured in Buda-100 stalagmite. Additionally, to support the model and results, the $^{87}\text{Sr}/^{86}\text{Sr}$ of local rain and the poorly developed lithosol above the cave are also included. The lithosol mostly consists of imperfectly weathered fragments of the host rock and, to a much lesser extent, of fine sediment of aeolian origin. In order to simulate as closely as possible, the behavior of rainwater seeping through the soil, a soil leaching procedure was made to quantify loss of soluble substances and colloids. Since the soil cover is thin (lithosol), this method was used as analogue of water infiltration after a rain event. The procedure was as follow. 100 g of soil were ground and mixed in order to obtain a homogeneous sample. 4 soil fractions with a soil:water ratio 1:1 (2 x 10mg:10mL and 2 x 30mg:30mL) were stirred for 24 hours. Then, they were centrifuged, and the supernatant was filtered through 2 microns for analysis.

Table S2.²³⁰Th dating results for stalagmite Buda-100. The error is 2σ error.

Sample Ref.	²³⁸ U (ppb)	²³² Th (ppt)	²³⁰ Th / ²³² Th (atomic x10 ⁻⁶)	d ²³⁴ U* (measured)	²³⁰ Th / ²³⁸ U (activity)	²³⁰ Th Age (yr) (uncorrected)	²³⁰ Th Age (yr BP) (corrected)	δ ²³⁴ U _{Initial} ** (corrected)
BUDA-0.1	133.0±0.2	292±6	379±8	132.7±2.4	0.0504±0.0003	4959±30	4841±50	135±2
BUDA-0.2	103.7±0.2	331±7	252±5	125.5±2.9	0.0489±0.0004	4837±38	4693±70	127±3
BUDA 0.3	106.1±0.2	505±10	156±3	119.1±3.1	0.0449±0.0003	4466±34	4281±94	121±3
BUDA 0.4	105.3±0.2	636±13	115±2	116.7±3.1	0.0422±0.0003	4197±35	3978±116	118±3
BUDA 1.1	89.6±0.2	650±13	95±2	107.6±1.7	0.0417±0.0007	4181±68	3929±151	109±2
BUDA 1.2	98.7±0.2	498±10	126±3	110.9±3.4	0.0385±0.0004	3840±41	3647±102	112±3
BUDA2-3	108.5±0.2	986±20	73±2	119.5±3.2	0.0393±0.0004	3896±47	3598±173	121±3
BUDA 3.4	115.1±0.3	903±18	73±2	123.7±4.1	0.0346±0.0003	3409±35	3145±148	125±4
BUDA 3.5	166.2±0.2	636±13	131±3	113.3±1.7	0.0305±0.0003	3027±26	2866±75	114±2
BUDA 3.6	108.8±0.3	729±15	71±2	129.3±4.2	0.0290±0.0003	2840±35	2607±127	130±4
BUDA 4.7	83.9±0.2	1076±22	37±1	132.1±4.8	0.0286±0.0004	2790±45	2400±237	133±5
BUDA 5.8	119.6±0.2	1298±26	37±1	120.8±2.3	0.0247±0.0003	2425±34	2082±202	121±2
BUDA 6.9	125.3±0.3	1808±36	27±1	116.5±2.7	0.0238±0.0003	2344±27	1907±268	117±3
BUDA 7.10	114.1±0.2	1558±31	19±0	112.0±3.1	0.0161±0.0002	1592±25	1173±254	112±3
BUDA 7.11	139.3±0.3	1241±25	23±1	113.2±3.4	0.0126±0.0003	1240±27	946±167	114±3

*δ²³⁴U = ([²³⁴U/²³⁸U]_{activity} - 1) × 1000. **δ²³⁴U_{initial} was calculated based on ²³⁰Th age (T), i.e., δ²³⁴U_{initial} = δ²³⁴U_{measured} × e^{-λ₂₃₄ × T}.Corrected ²³⁰Th ages assume the initial ²³⁰Th/²³²Th atomic ratio of 4.4 ± 2.2 × 10⁻⁶. Those are the values for a material at secular equilibrium, with the bulk earth ²³²Th/²³⁸U value of 3.8. The errors are arbitrarily assumed to be 50%.

BP stands for "Before Present" where the "Present" is defined as the year 1950 A.D.

²³⁰Th dating results for stalagmite Buda-100. ²³⁰Th age-dates were obtained using multicollector inductively coupled plasma–mass spectrometry (ICP-MS), following methods described in (88) and (89). The error is 2σ error.

Caption for Excel file F1 (abe6102_Excel_F1.xlsx)

This worksheet summarizes the mass-balance model used for calculating past dust deposition rates ($\text{g}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$) in the study region (Kaite Cave, N Spain) from $^{87}\text{Sr}/^{86}\text{Sr}$ ratios measured in speleothem calcite. See Material and Methods section in the article for details.

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