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Microbial life and biogeochemical cycling on land 3,220 million years ago

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SUPPLEMENTARY INFORMATION TO

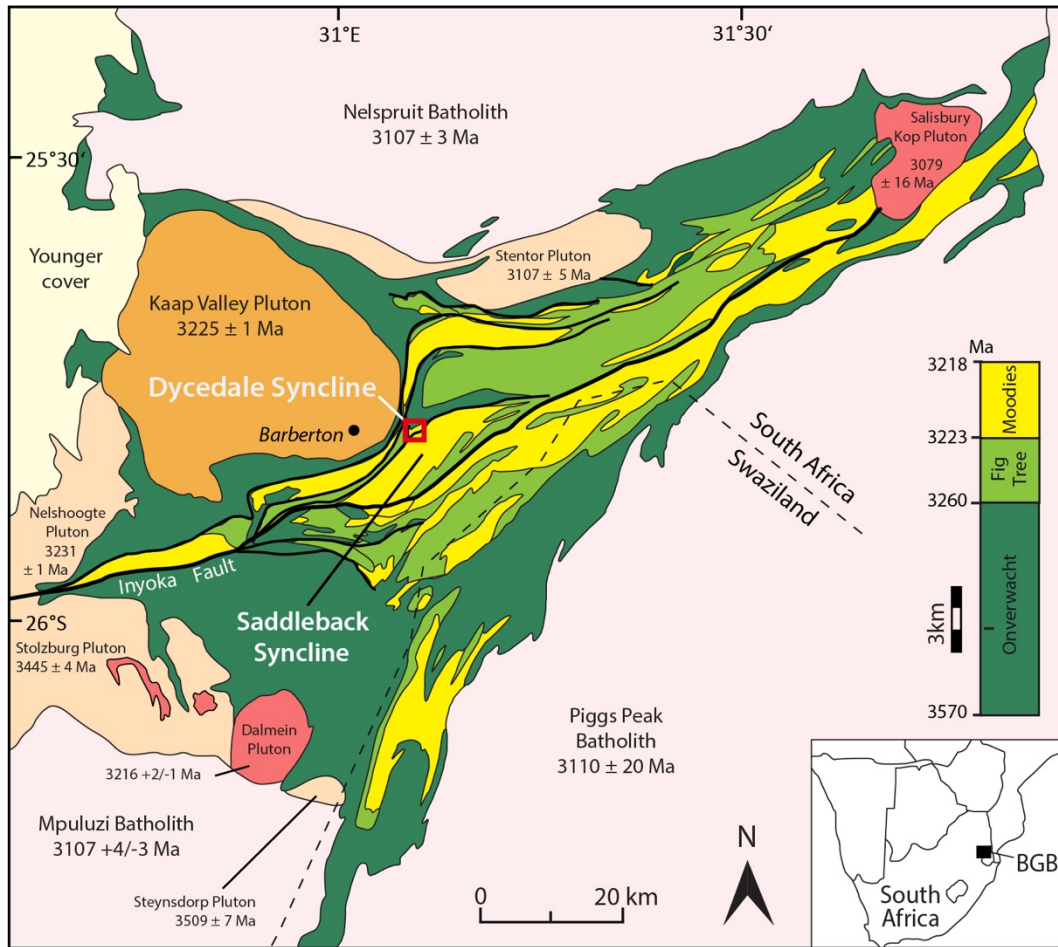
Microbial life and biogeochemical cycling on land 3,220 million years ago

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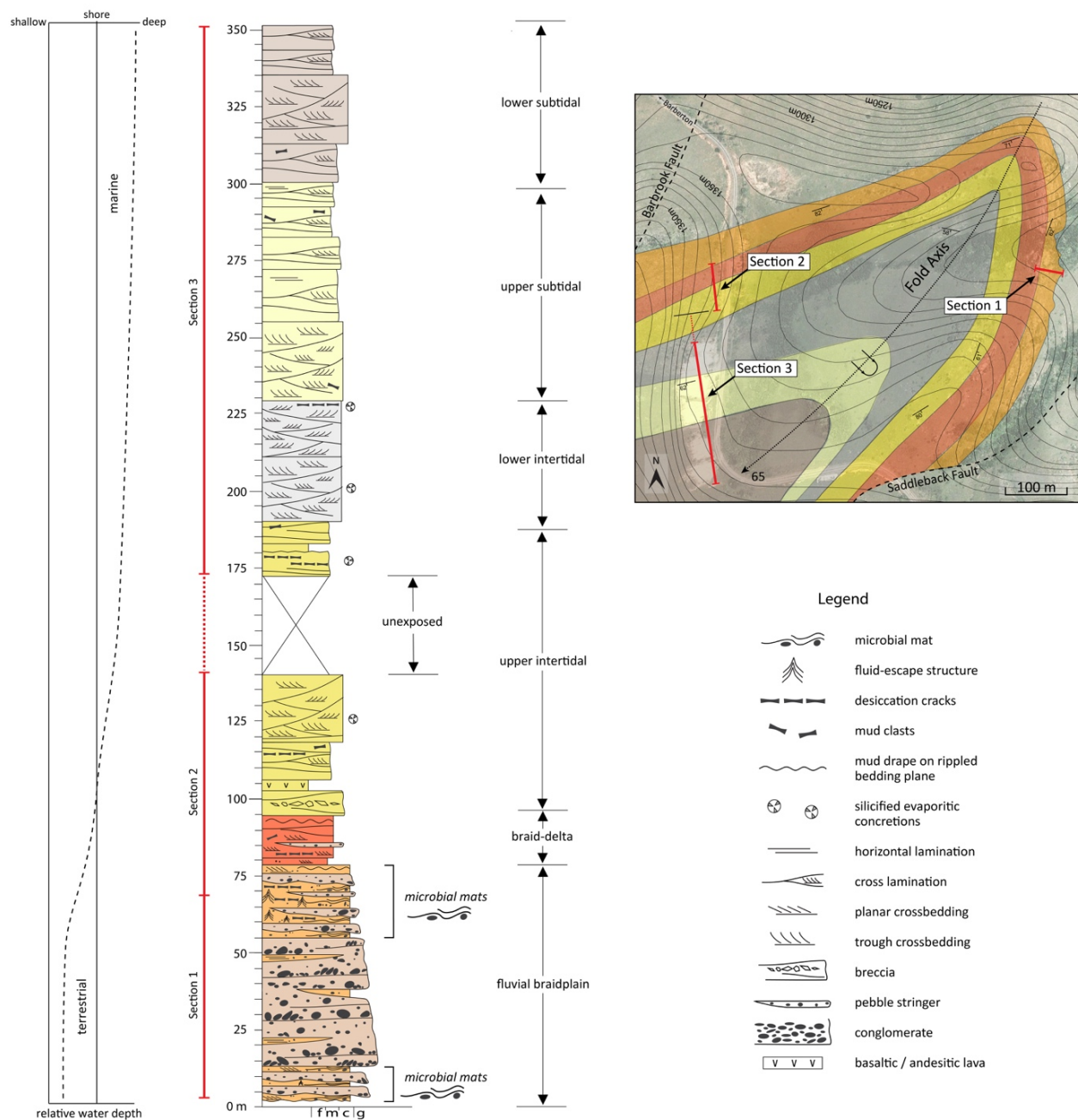
Evidence for terrestrial life in the Archean

In the literature the term “terrestrial” can have multiple meanings as pointed out by Labandeira¹ and Berladi-Campesi². In a broader context “terrestrial” is used to refer to land, the planet Earth in general, and non-aquatic environments. In a closer context, some terrestrial environments can be fully aquatic ecosystems (e.g. lakes, ponds, and geothermal springs), whereas others frequently experience periods of subaerial exposure and desiccation (e.g. fluvial systems, alluvial fans, wetlands, peats, and playas). In the context of our manuscript we define “terrestrial” as a clearly non-marine, aquatic paleoenvironment, which experienced periodically subaerial exposure and desiccation. In detail we consider any life on the emerged continental surface, aquatic or subaerial, as terrestrial.

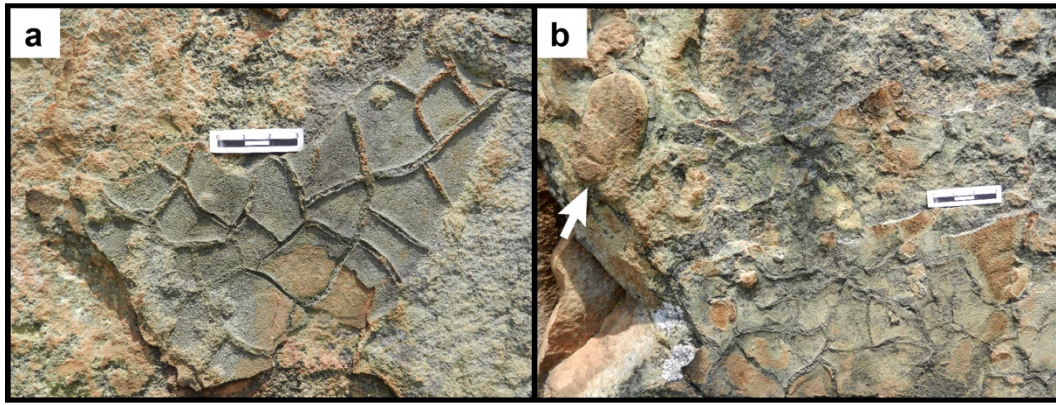
Fossil evidence for the microbial colonization of terrestrial habitats during Archean times is extremely sparse^{2–4}. Until recently, the oldest known macroscopically-visible traces were ~2,700 Ma old fluvio-lacustrine stromatolites preserved in the Ventersdorp Supergroup (South Africa)⁵ and the Fortescue Group (Tumbiana Formation, Australia)^{6–8}. The 3,220 Ma terrestrial microbial mats of the Moodies Group (BGB, South Africa) described in the present study now extend the fossil record of life on land by ~500 million years into near-coastal but clearly fluvial aquatic settings. Geochemical traces preserved in Moodies Group paleosols provide additional evidence for the presence of a Paleoarchean terrestrial biosphere^{9,10}. The Moodies Group terrestrial mats of the Dycedale Syncline are similarly well preserved as fossil marine microbial mats outcropping in tidal deposits of the adjacent Saddleback Syncline (approx. 4 km south of the Dycedale Syncline (Supplementary Fig. 1), which have a well-established biogenicity^{11–14}.



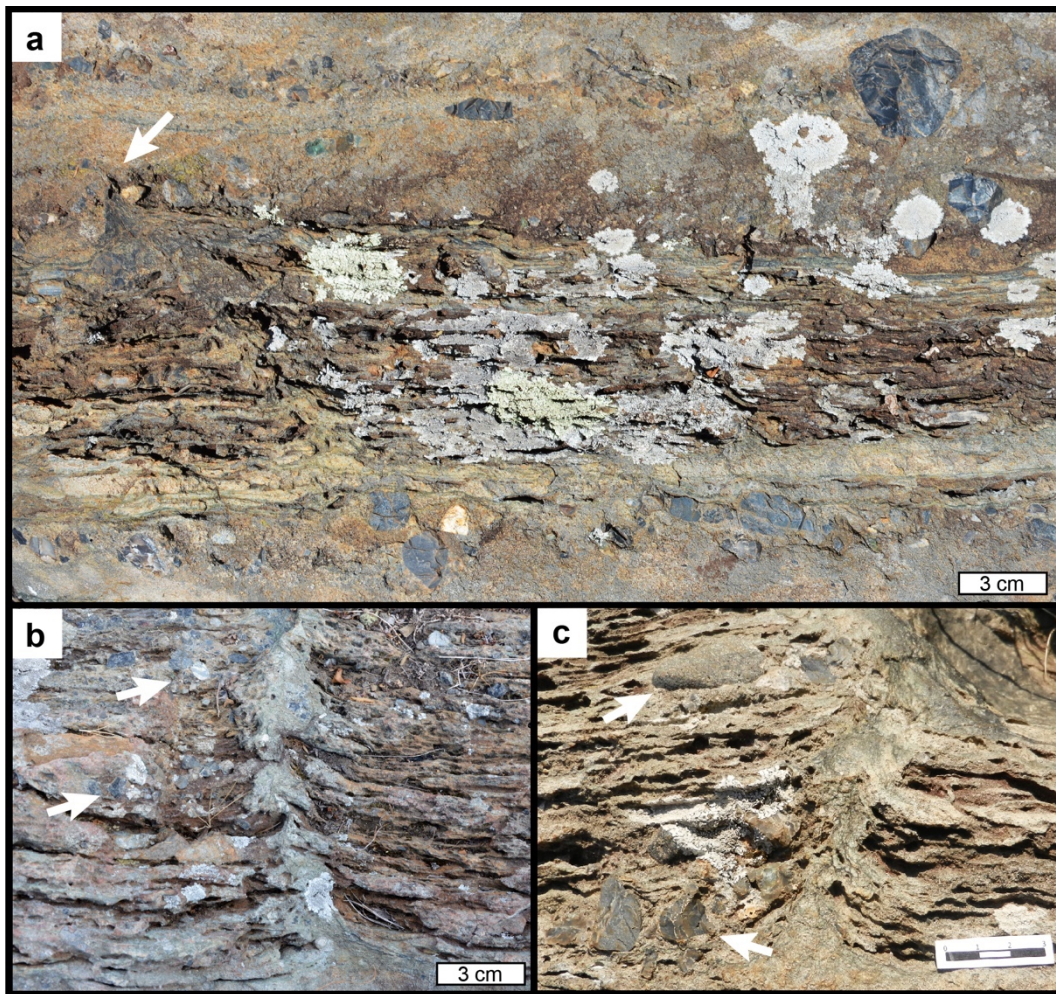
Supplementary Figure 1. Simplified geological map of the Barberton Greenstone Belt (BGB) of South Africa, Swaziland, and its surrounding plutons. Fossilized terrestrial microbial mats are preserved in fluvial sandstones of the Moodies Group in the Dycedale Syncline, a small triangular-shaped syncline in the central part of the BGB. Marine microbial mats are widespread in tidal sandstones outcropping in the adjacent Saddleback Syncline¹³.



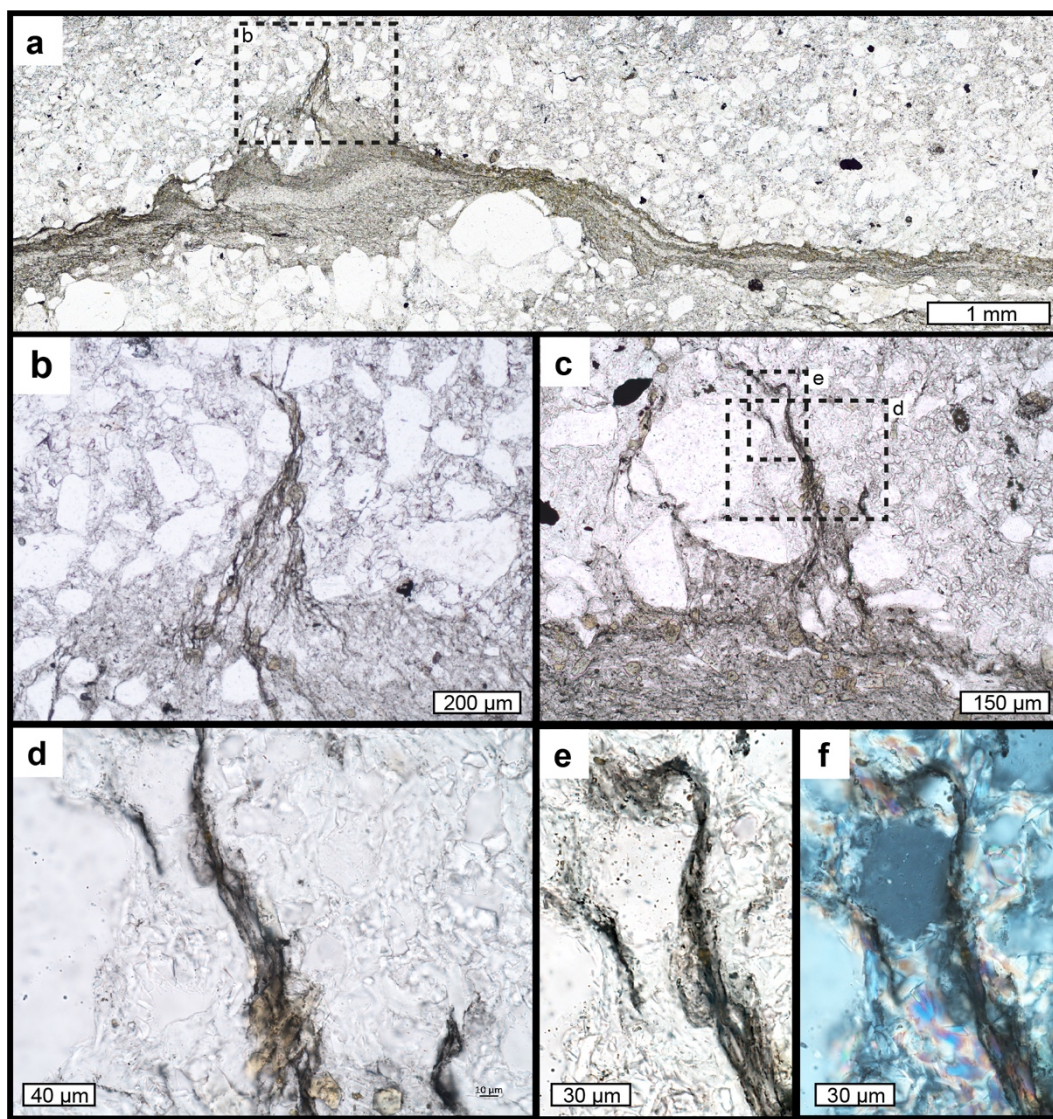
Supplementary Figure 2. Composite stratigraphic column of the Dycedale Syncline. The positions of the measured sections are shown in the geological map in the upper right. Note that microbial mats are restricted to the basal terrestrial part of the succession and are confined to gravelly sandstones interbedded with fluvial-alluvial conglomerates. They are successively overlain by both tidally-influenced, and subtidal siliciclastic deposits^{15,16}.



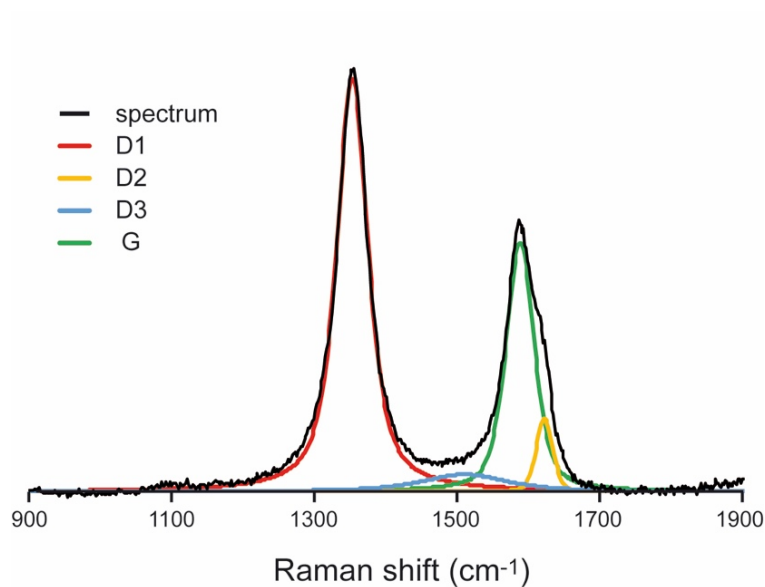
Supplementary Figure 3. a, b, Field photographs (bedding plane view) of desiccation cracks in thin mudstone layers that occur interbedded with the terrestrial, mat-bearing, gravelly sandstones (arrow).



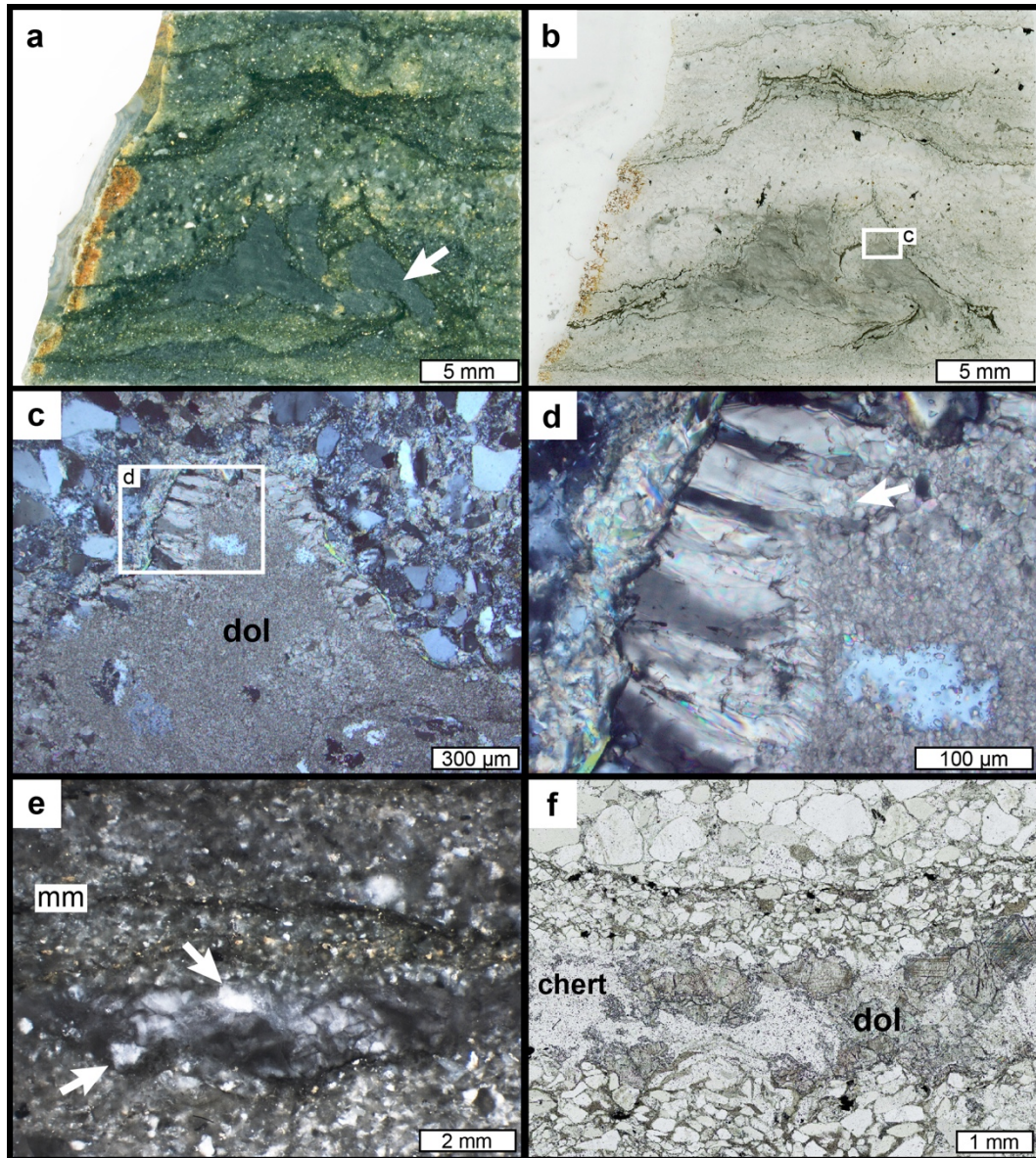
Supplementary Figure 4. Field photographs (cross-section view) of mat-associated fluid-escape structures that were likely triggered by a rapid overburden of sediment consistent with high sedimentation rates in fluvial-alluvial environments. **a,** Subvertical fluid-escape structure (arrow) disrupting a thin interval of microbial mats, which is in turn buried by gravelly sandstones. **b, c,** Upward-curved mats and pebbles bordering a central dewatering channel (arrows). Note that the force of the escaping fluids must have been strong enough to transport large pebbles laterally within the fluidized sediment towards the central conduit while dragging multiple layers of cohesive mats upwards into the conduit.



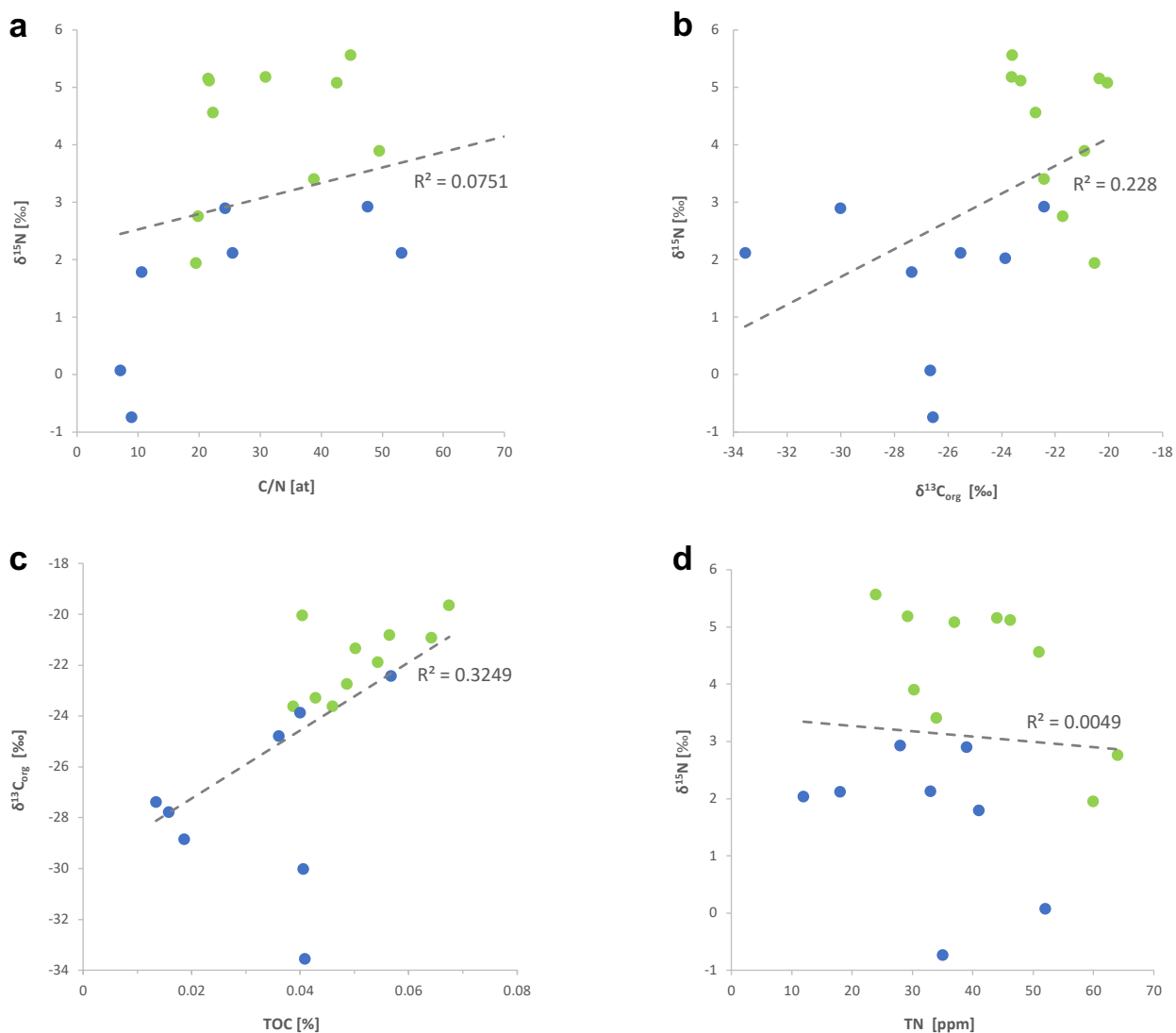
Supplementary Figure 5. Transmitted (a-e) and polarized (f) light photomicrographs of terrestrial microbial mat remnants from the Dycedale Syncline. **a**, Thick microbial mat with well-preserved kerogenous laminae draping coarse sand grains. **b**, Partially eroded mat laminae show bundled of interwoven filamentous microstructures (close-up view of the framed area in (a)). **c**, Microbial laminae draping detrital sediment particles in the upper part of the mat. **d**, **e**, **f**, Close-up views of the framed areas in (c) showing cohesive networks of bundled filamentous structures.



Supplementary Figure 6. Raman spectrum of carbonaceous material from sample 14-452-1B2 of the Moodies Group. Peak-decomposition is shown for a four-peak fit, assigning a D1, D2, D3-, and G-peak at ca. 1350 cm⁻¹, 1620 cm⁻¹, 1500 cm⁻¹, and 1600 cm⁻¹, respectively. (see ref.¹⁷ for detailed description). In all obtained spectra a D4-peak at ca. 1200 cm⁻¹ was absent and therefore not assigned during the peak decomposition procedure.



Supplementary Figure 7. Polished slab and thin section photomicrographs of cavity-filling dolomitic cements associated with the terrestrial (a-d) and marine mats (e, f). **a, b**, Deformed and partially ruptured terrestrial microbial mats due to precipitation of early diagenetic dolomite cements (arrow). **c, d**, Close-up view of the dome showing inward-facing bladed (arrow) to granular dolomite (dol) cements. **e, f**, Marine microbial mat (mm) underlain by bedding-parallel, partly silicified cavity with blocky dolomite remnants (arrows).



Supplementary Figure 8. Plots of organic carbon and nitrogen isotopic data of terrestrial (green) and marine (blue) microbial mats. **a**, Nitrogen isotopes ($\delta^{15}\text{N}$) plotted against ratios of organic carbon to total nitrogen (C/N). **b**, Nitrogen isotopes ($\delta^{15}\text{N}$) versus organic carbon isotopes ($\delta^{13}\text{C}_{\text{org}}$), **c**, Organic carbon isotopes ($\delta^{13}\text{C}_{\text{org}}$) versus total organic carbon (TOC) **d**, Nitrogen isotopes ($\delta^{15}\text{N}$) versus total nitrogen content (TN). No strong trends are observed with the exception of a possible trend in $\delta^{13}\text{C}_{\text{org}}$ vs. TOC. This trend is opposite to that expected for post-depositional volatilization of isotopically light carbon, suggesting that it is not a diagenetic artifact, but rather a primary feature of the microbial mats.

Supplementary Table 1 | Raman parameters and calculated metamorphic temperatures for carbonaceous laminae from terrestrial and marine mats from the Moodies Group

Analysis	D1-FWHM	Dt-FWHM	IDt/IGt	R1	R2	T-R2 (°C)*	T-D1 (°C)†	Paleoenvironment
13-452-1B2-spec1a	60.98	63.24	1.41	1.46	0.60	373	347	terrestrial
13-452-1B2-spec2a	57.59	57.43	1.49	1.52	0.62	366	354	terrestrial
13-452-1B2-spec3a	54.05	52.17	1.58	1.59	0.61	368	362	terrestrial
13-452-1B2-spec4a	53.40	51.67	1.62	1.60	0.62	364	363	terrestrial
13-452-1B2-spec5a	51.80	51.54	1.34	1.30	0.58	384	367	terrestrial
13-452-1B2-spec6a	63.64	61.27	1.53	1.51	0.63	362	341	terrestrial
13-452-1B2-spec8a	56.95	55.35	1.48	1.49	0.61	371	356	terrestrial
13-452-1B2-spec10a	51.71	50.45	1.39	1.34	0.58	382	367	terrestrial
13-452-1B2-spec11a	52.60	52.31	1.64	1.66	0.63	359	365	terrestrial
13-452-1B2-spec13a	57.29	54.92	1.46	1.42	0.60	375	355	terrestrial
Average	56.00	55.03	1.49	1.49	0.61	370	358	terrestrial
Stdev	4.03	4.37	0.10	0.11	0.02	8	8	
14-452-1B9-spec2a	58.06	58.10	1.63	1.69	0.63	362	353	terrestrial
14-452-1B9-spec4a	57.05	57.54	1.40	1.41	0.60	375	355	terrestrial
14-452-1B9-spec5a	59.94	60.44	1.48	1.52	0.61	367	349	terrestrial
14-452-1B9-spec6a	57.18	56.62	1.24	1.23	0.57	386	355	terrestrial
Average	58.06	58.18	1.44	1.46	0.60	372	353	terrestrial
Stdev	1.34	1.63	0.16	0.19	0.02	10	3	

* Beyssac et al.¹⁸

† Kouketsu et al.¹⁹

Analysis	D1-FWHM	Dt-FWHM	IDt/IGt	R1	R2	T-R2 (°C)*	T-D1 (°C)†	Paleoenvironment
11-196-4-spec1a	60.71	62.14	1.34	1.37	0.60	375	347	marine
11-196-4-spec2a	61.19	61.67	1.46	1.46	0.62	365	346	marine
11-196-4-spec3a	55.48	58.65	1.39	1.45	0.61	371	359	marine
11-196-4-spec4a	57.18	56.44	1.58	1.59	0.62	366	355	marine
11-196-4-spec5a	60.73	62.00	1.51	1.53	0.62	365	347	marine
11-196-4-spec6a	57.19	58.27	1.43	1.43	0.60	375	355	marine
11-196-4-spec7a	63.97	61.89	1.59	1.63	0.64	356	340	marine
Average	59.49	60.15	1.47	1.50	0.61	368	350	marine
Stdev	2.96	2.32	0.09	0.09	0.02	7	6	
13-441-B5-spec1a	48.72	49.59	1.39	1.37	0.59	379	373	marine
13-441-B5-spec2a	48.37	47.54	1.66	1.62	0.62	367	374	marine
13-441-B5-spec3a	47.57	47.12	2.20	2.24	0.68	337	376	marine
13-441-B5-spec4a	49.49	50.41	1.78	1.79	0.64	357	372	marine
13-441-B5-spec5a	50.21	50.09	1.71	1.68	0.62	364	370	marine
13-441-B5-spec6a	41.27	43.23	1.21	1.15	0.55	397	389	marine
13-441-B5-spec7a	44.62	43.08	1.18	1.12	0.55	397	382	marine
Average	47.18	47.29	1.59	1.57	0.61	371	377	marine
Stdev	3.16	3.09	0.36	0.39	0.05	22	7	

* Beyssac et al.¹⁸

† Kouketsu et al.¹⁹

Supplementary Table 2 | Inorganic carbon and oxygen isotope data of mat-associated early diagenetic carbonates

Sample	$\delta^{13}\text{C}_{\text{carb}}$ [‰]	$\delta^{18}\text{O}_{\text{carb}}$ [‰]	Paleoenvironment
14-452-1B-1	0.46	-16.07	terrestrial
14-452-1B-2	0.46	-16.00	terrestrial
14-452-1B-3	0.46	-16.05	terrestrial
14-452-1B-4	0.49	-16.04	terrestrial
14-452-1A-1	0.63	-15.04	terrestrial
14-452-1A-2	0.56	-15.40	terrestrial
14-452-1A-3	0.46	-16.48	terrestrial
14-452-1A-4	0.93	-14.21	terrestrial
14-452-1A-5	0.66	-14.98	terrestrial
14-452-1A-6	0.42	-15.61	terrestrial
12-303-3C1	-0.51	-15.63	marine
12-303-3C2	-0.47	-14.80	marine
12-303-3C3	-0.45	-15.60	marine
12-303-3C4	-0.16	-14.99	marine
12-303-3C5	-0.80	-15.03	marine
12-303-3C6	-0.28	-15.22	marine

Supplementary Table 3 | Organic carbon isotope data for all measured terrestrial and marine microbial mat samples of the Moodies Group

Sample	$\delta^{13}\text{C}_{\text{org}} [\text{‰}]$	Paleoenvironment
12-322-1	-23.29	terrestrial
12-322-2	-22.75	terrestrial
13-431-1	-23.18	terrestrial
13-431-2	-21.73	terrestrial
14-451-1B-4A	-17.85	terrestrial
14-451-1B-4A rpt	-19.03	terrestrial
14-451-1B-4A rpt	-20.04	terrestrial
14-451-1B-4A rpt	-20.55	terrestrial
14-451-1B-4B	-20.05	terrestrial
14-451-1B-4B rpt	-19.37	terrestrial
14-451-1B-4B rpt	-20.80	terrestrial
14-452-1B	-22.88	terrestrial
14-452-1B rpt	-21.61	terrestrial
14-452-2C	-21.21	terrestrial
14-452-2C rpt	-21.02	terrestrial
14-452-2C rpt	-21.34	terrestrial
14-452-2C rpt	-20.54	terrestrial
14-452-3B	-21.47	terrestrial
14-452-3B rpt	-22.51	terrestrial
14-452-3B rpt	-22.48	terrestrial
14-452-3B rpt	-23.23	terrestrial
14-452-4C	-20.36	terrestrial
14-452-4C rpt	-20.61	terrestrial
14-452-4C rpt	-20.10	terrestrial
14-452-1B-1B	-20.07	terrestrial
14-452-1B-2B	-18.55	terrestrial
14-452-1B-3B	-18.81	terrestrial
14-452-1B-4B	-20.51	terrestrial
14-452-1B-5A	-23.46	terrestrial
14-452-1B-4B	-21.67	terrestrial
17-467	-22.51	terrestrial
17-468	-23.63	terrestrial
17-469	-23.62	terrestrial
17-470	-20.93	terrestrial
17-471	-22.36	terrestrial
17-472	-22.28	terrestrial
11-034	-33.56	marine
11-196-2	-33.90	marine
11-196-3	-26.67	marine
11-262-1	-33.41	marine
11-262-2	-25.54	marine
12-304	-27.36	marine
12-314	-25.68	marine
12-323-2	-22.43	marine
12-332-1	-31.41	marine
12-332-2	-25.49	marine
13-441	-26.57	marine
13-444_A	-23.88	marine
13-441-M	-25.39	marine
13-441-M1A	-30.42	marine
13-441-M1B	-32.03	marine
13-441-M2A	-26.69	marine
13-441-M2B	-27.84	marine
13-441_A1-1*	-23.72	marine
13-441_A1-2*	-26.18	marine
13-441_A1-3*	-29.65	marine
13-441_A1-4*	-22.29	marine
13-441_A1-5*	-26.04	marine
13-441_A1-6*	-24.25	marine
13-441_A2-1*	-30.82	marine
13-441_A2-2*	-21.32	marine
13-441_A2-3*	-24.68	marine
13-441_A2-4*	-27.81	marine
13-441_A2-5*	-32.34	marine
13-441_A2-6*	-29.39	marine
17-458	-24.82	marine

rpt repeated measurement

* SIMS data from Homann et al.¹⁴

Supplementary Table 4 | Organic carbon and total nitrogen isotope data for all measured terrestrial and marine microbial mat samples of the Moodies Group

Sample	$\delta^{13}\text{C}_{\text{org}}$ [‰]	TOC [%]	$\delta^{15}\text{N}$ [‰]	SD [‰]	TN [ppm]	TOC/TN [%]	C/N [atm.]	Paleoenvironment
12-322-1	-23.29	0.04	5.12	0.08	46	9.27	21.63	terrestrial
12-322-2	-22.75	0.05	4.57	0.11	51	9.53	22.24	terrestrial
13-431-2	-21.73	0.05	2.76	0.14	64	8.49	19.81	terrestrial
14-452-1B1	-20.06	0.07	5.08	0.19	37	18.23	42.54	terrestrial
14-452-1B3	-22.42	0.06	3.41	0.09	34	16.61	38.76	terrestrial
14-452-1B4	-20.36	0.04	5.15	0.05	44	9.19	21.44	terrestrial
14-452-1B5	-20.54	0.05	1.95	0.13	60	8.36	19.51	terrestrial
17-468	-23.63	0.04	5.19	0.07	29	13.25	30.91	terrestrial
17-469	-23.62	0.05	5.56	0.12	24	19.19	44.77	terrestrial
17-470	-20.93	0.06	3.90	0.09	30	21.24	49.55	terrestrial
11-034-2	-33.56	0.04	2.53	0.23	16	26.11	60.93	marine
11-034-2 rpt	-33.56	0.04	1.71	0.06	21	19.48	45.44	marine
11-034-2 avg	-33.56	0.04	2.12	0.14	18	22.79	53.19	marine
11-196-3	-26.67	0.02	0.07	0.08	52	3.04	7.10	marine
11-262-2	-25.54	0.04	2.12	0.32	33	10.94	25.52	marine
12-304	-27.36	0.02	1.79	0.11	41	4.55	10.61	marine
12-323-2	-22.43	0.06	3.12	0.19	27	20.91	48.78	marine
12-323-2 rpt	-22.43	0.06	2.74	0.09	29	19.88	46.38	marine
12-323-2 avg	-22.43	0.06	2.93	0.14	28	20.39	47.58	marine
13-441	-26.57	0.01	-0.74	0.17	35	3.84	8.96	marine
13-441_M1	-30.02	0.04	2.90	0.18	39	10.40	24.27	marine
13-444_A	-23.88	0.04	2.03	0.14	12	33.41	77.96	marine

rpt repeated measurement
avg average

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