Supplementary Material for Manuscript "Open ocean vs. continentally-derived iron cycles along the Neoarchean Campbellrand-Malmani carbonate platform, South Africa"

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Additional method section: X-ray Diffraction (XRD)

XRD analyses were conducted in the Materials Analysis and Research Laboratory (MARL) of Iowa State University. For this study representative samples for 'pure' carbonates, silicified carbonates, and mudrocks were measured on a Siemens D 500 diffractometer using CuKα radiation. Powdered samples were placed in a specimen holder, covered with plexiglass, and measured under conditions of 45 kV and 30 mA. XRD patterns were acquired over a 2θ range of 4 to 75° and subsequently analyzed using JADE software. In this study representative carbonate samples were analyzed by XRD to complement results of XANES analyses. Pure carbonate samples clearly show a varying mixture of dolomite and quartz, depending on the degree of silicification, with minor amounts of calcite (fig. 4). However, no Fe-mineral phases could be detected by XRD, probably because the detection limit of this method for crystalline material is between ~ 0.1 and 0.5 wt-%. Most of the samples investigated here have Fe concentrations within this range. Thus, XANES analyses are necessary to determine the Fe phase and speciation within the carbonates.

Additional Figures

Figs. S1 a-d: Thin section images and XAS maps of edge-jump and edge-position of carbonate and mudrock samples from KMF-5 and BH-1, which were analyzed under fluorescence or transmission mode. Dark blue areas in fluorescence or transmission maps represent excluded spectra, which were over- or under-saturated or had a poor signal-to-noise ratio. Linear combination fitting (LCA fit, red line) was performed on representative spectra (blue line). Mineral proportions are indicated below those fits. Thin section images analyzed under transmission mode are representative for the chips that were actually analyzed, because petrographic details are more visible in the thin sections.

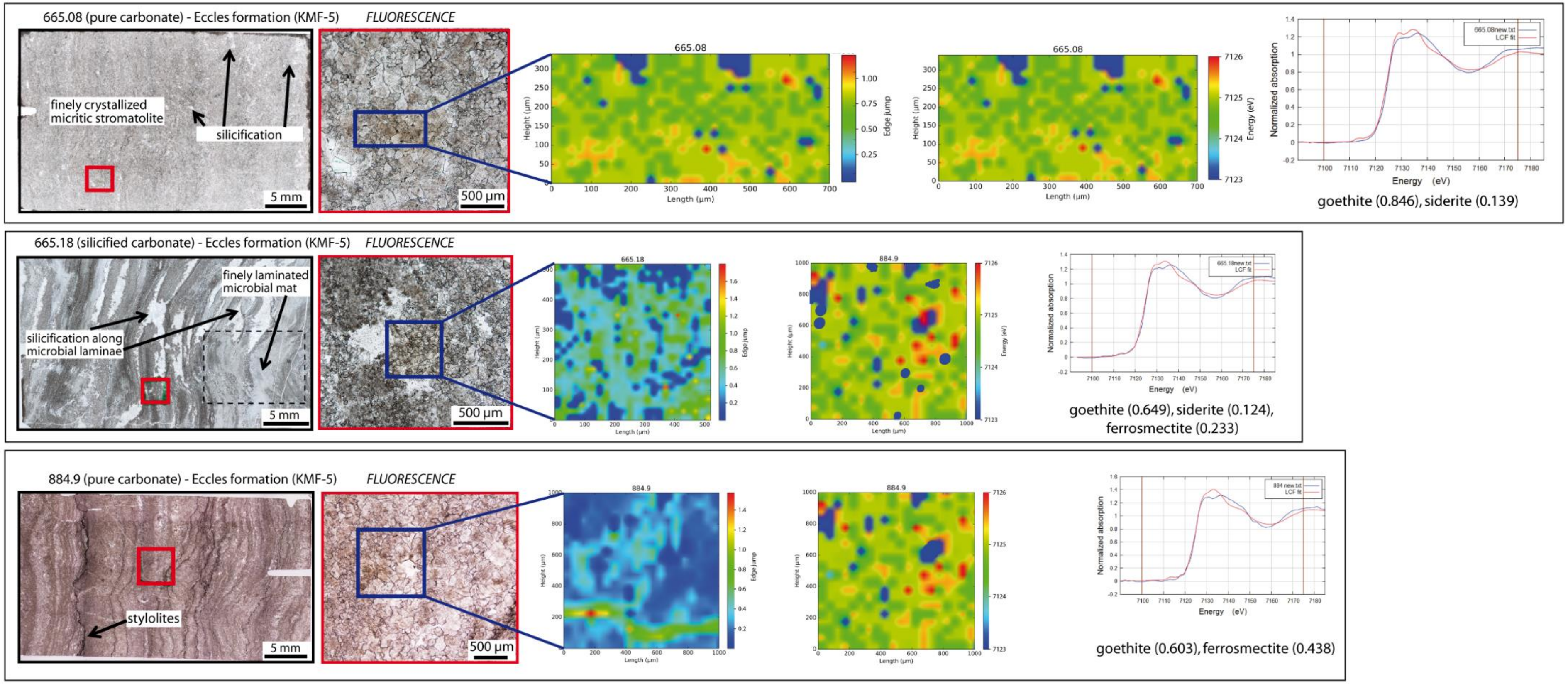

Fig. S1 a)

665.08 (KMF-5): Micritic laminated stromatolite with small cavities filled with silica, which are of lighter color than the surrounding matrix; blue box marks the analyzed section visible in the edge jump and energy maps

665.18 (KMF-5): Like sample 665.08, but heavily silicified. Migration of the silica appears unidirectional, following intercrystalline pores, presumably from stromatolitic carbonate layers and from cracks. Silicification occurred during early diagenesis, and reflects peritidal conditions.

884.9 (KMF-5): Laminated fine-grained carbonate with stylolites that originate from pressure dissolution after carbonate formation.

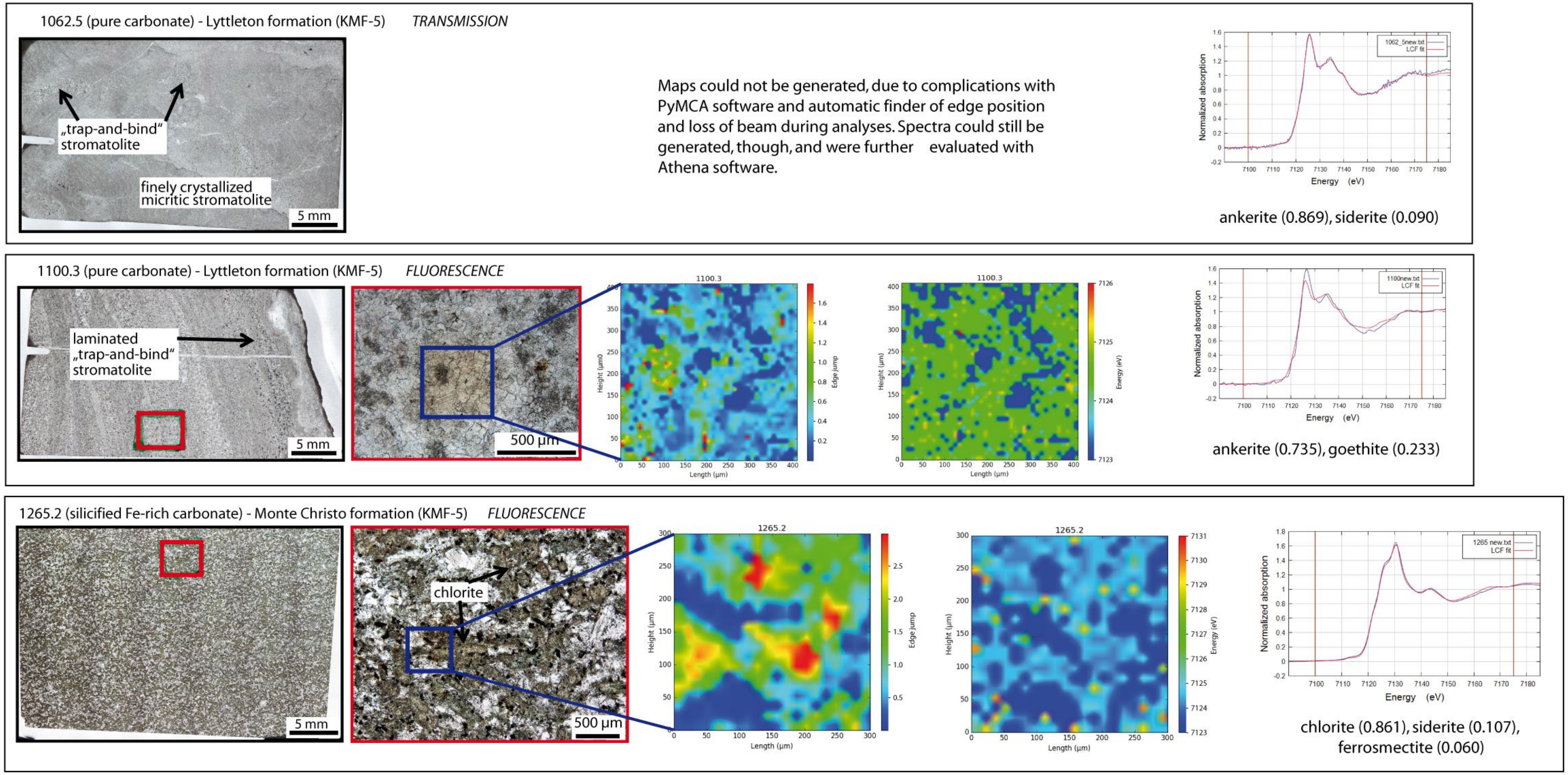

Fig. S1 b)

1062.5 (KMF-5): Fine microbial carbonate with small peloid-like carbonate debris trapped and bound by microbial laminae.

1100.3 (KMF-5): Laminated fine-grained agglutinated stromatolite. Small peloids are of microbial origin and are typical for trapping and binding of particles (mostly carbonate debris) in the microbial mats. This represents wave-agitated facies between shallow subtidal to intertidal conditions.

1265.2 (KMF-5): Fe-rich, silicified carbonates, containing siliciclastics, in particular clay minerals (i.e. ferrosmectite), chlorite, and opaque minerals. Probably related to deposition of Kamden Member in GWA.

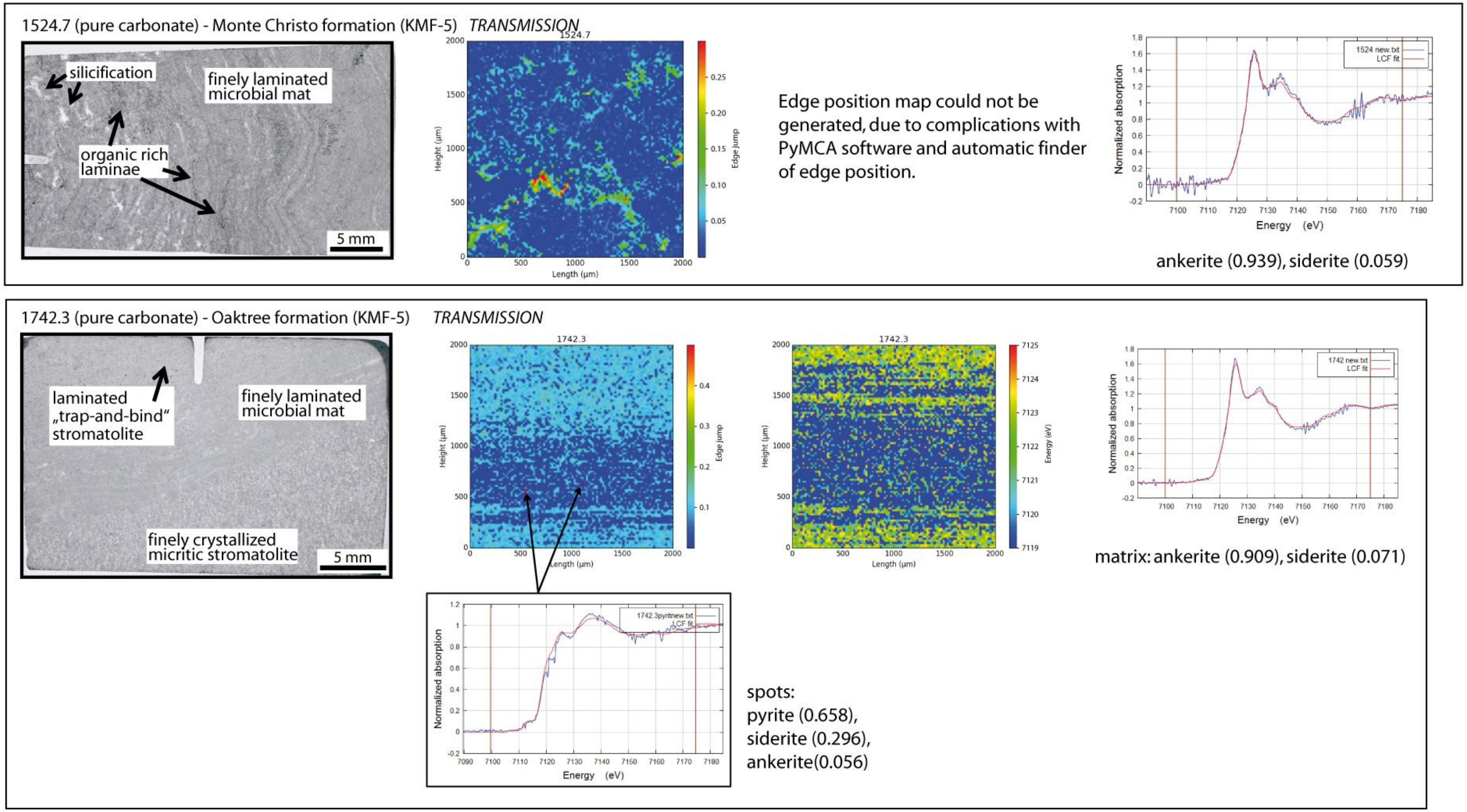

Fig. S1 c)

1524.7 (KMF-5): Laminated fine-grained carbonate, with small silica-filled cavities and organic-rich layers. Laminated fine-grained carbonate with small peloid-like carbonate debris trapped and bound by microbial laminae.

1742.3 (KMF-5): Laminated and fine microbial carbonate, containing some “trap and bind” structure, indicative for shallow subtidal conditions.

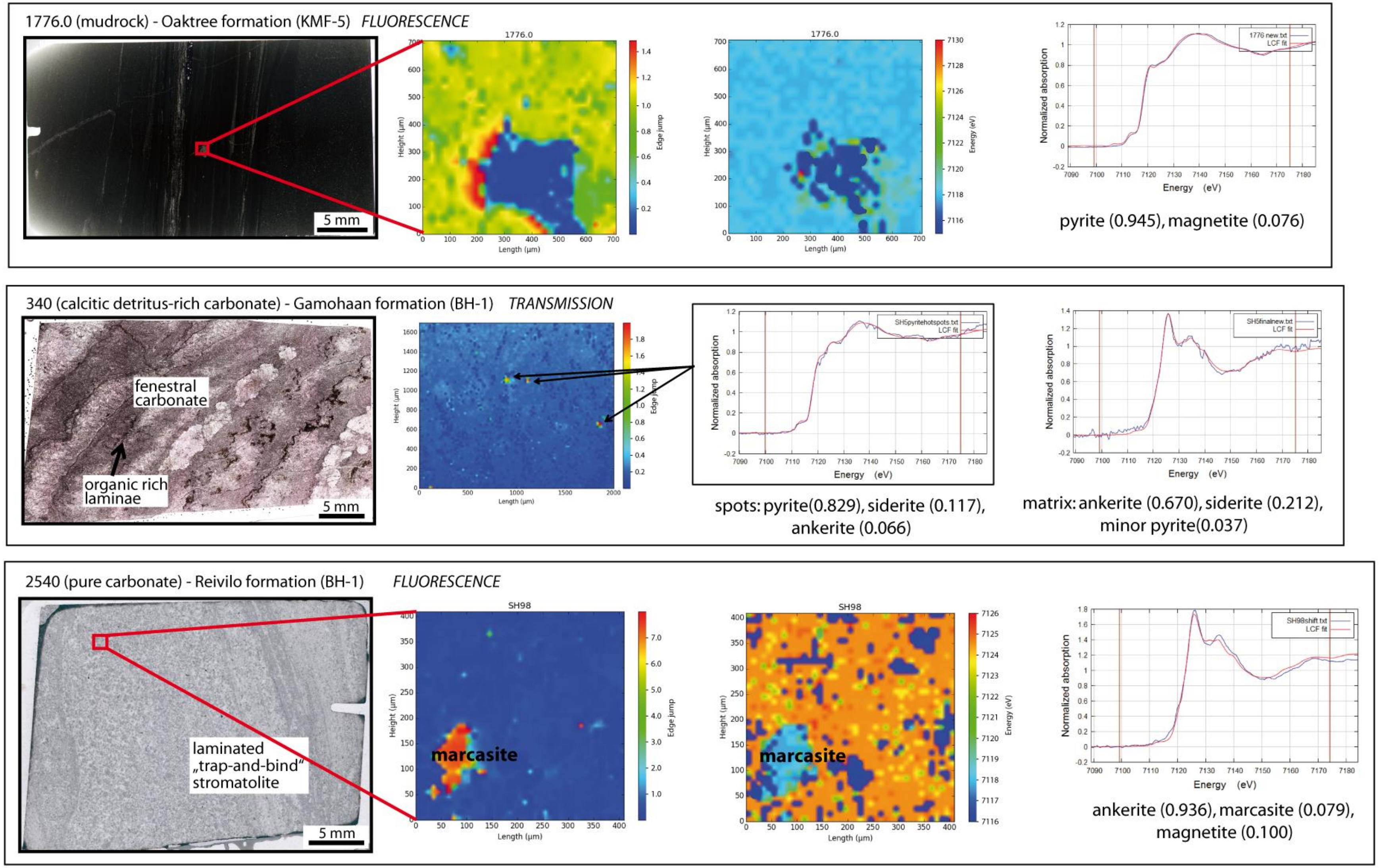

Fig. S1 d)

1776.0 (KMF-5): Organic and siliciclastic rich mudrock, very fine grainsize, containing some Fe-sulfides.

340 (BH-1): Alternating dark and light laminated carbonate layers, finely crystallized with coarsely crystalline fenestral structures. Depositional conditions were intertidal to lagoonal.

2540 (BH-1): Laminated and fine microbial carbonate, containing some “trap and bind” structure, indicative for shallow subtidal conditions.

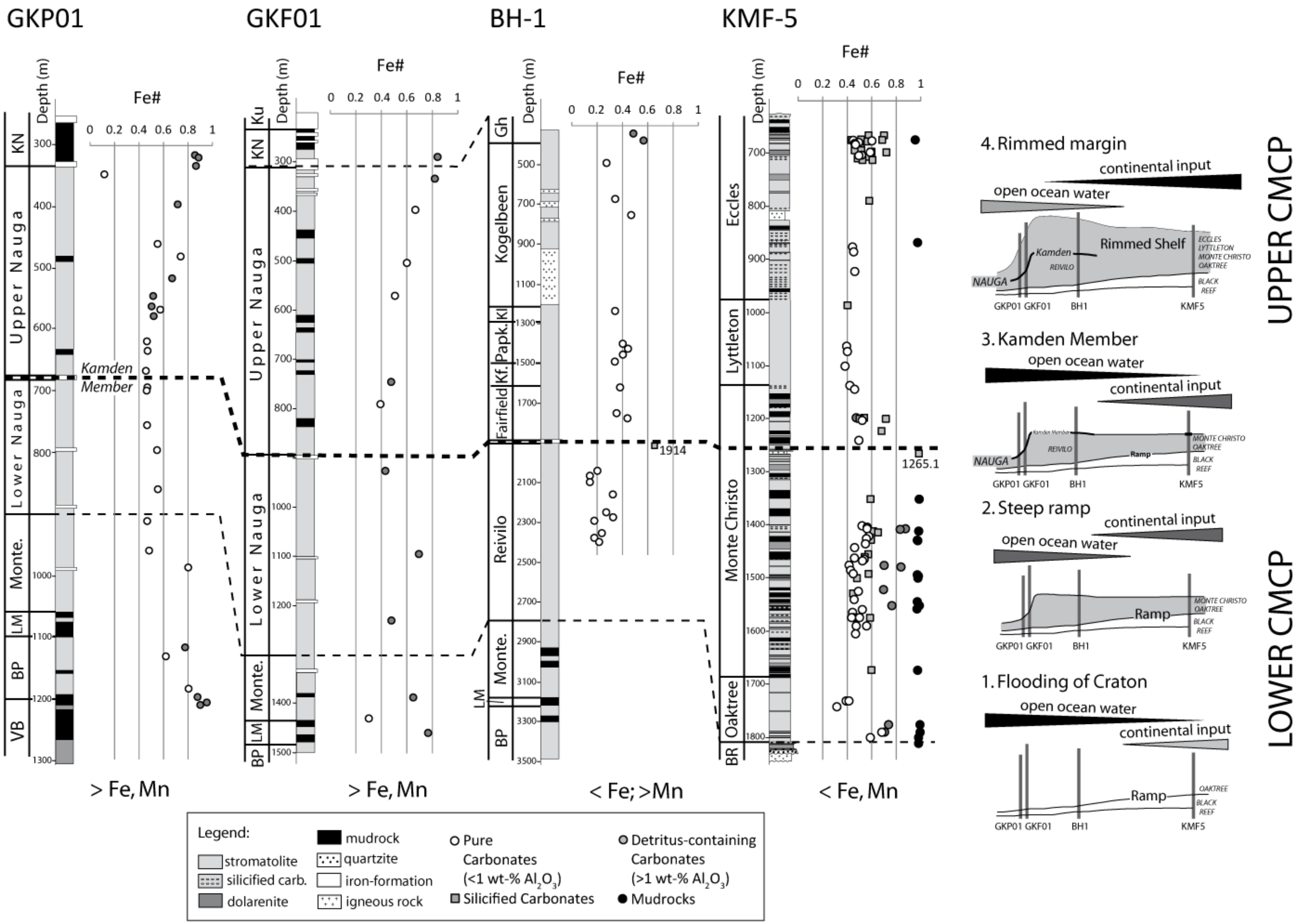

Fig. S2 a)

Fe# = ([Fe_{tot}/(Fe_{tot}+Mn_{tot})]) of slope and shelf sediments reveal a dependence on water depth and source water influx. Data of GKP01 and GKF01 are from Voegelin et al. (2010). BP-Boomplaas; LM-Lokamonna; Monte Monteville; KN-Klein Naute; Ku-Kuruman; Fairfi.-Fairfield; Kf.-Klipfonteinheuwel; Papk.-Papkuil; K.-Klippan; Gh-Gamohaan; BR-Black Reef). Modified from Eroglu et al. (2017).

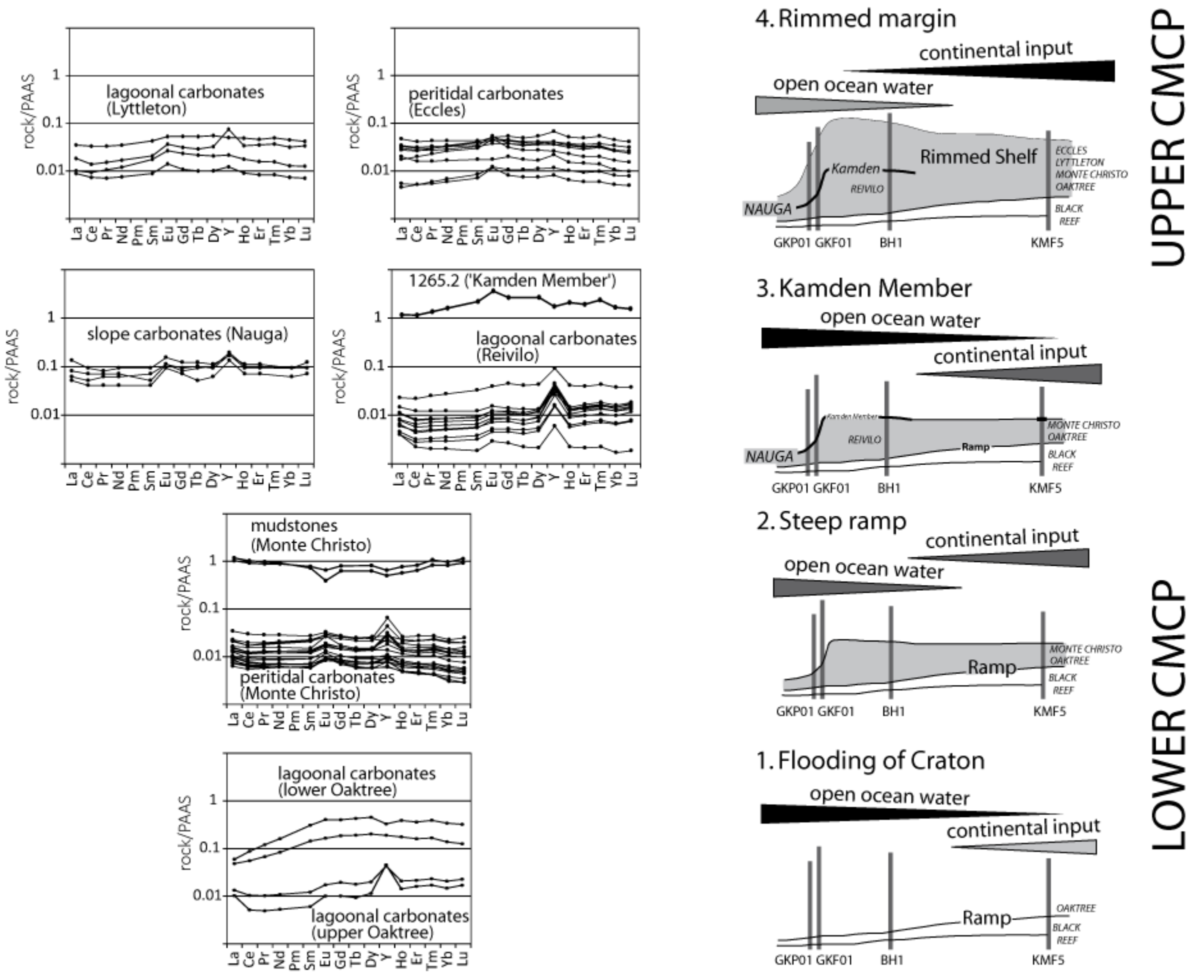

Fig. S2 b)

REE+Y signatures of slope and shelf sediments reflect open ocean water and source water signals. Data of GKP01 and GKF01 are from Voegelin et al. (2010). BP-Boomplaas; LM-Lokamonna; Monte Monteville; KN-Klein Naute; Ku-Kuruman; Fairfi.-Fairfield; Kf.-Klipfonteinheuwel; Papk.-Papkuil; K.-Klippan; Gh-Gamohaan; BR-Black Reef). Concentrations are normalized to post-Archean Australian Shale (PAAS, Taylor and MacLennan, 1985). Modified from Eroglu et al. (2017).

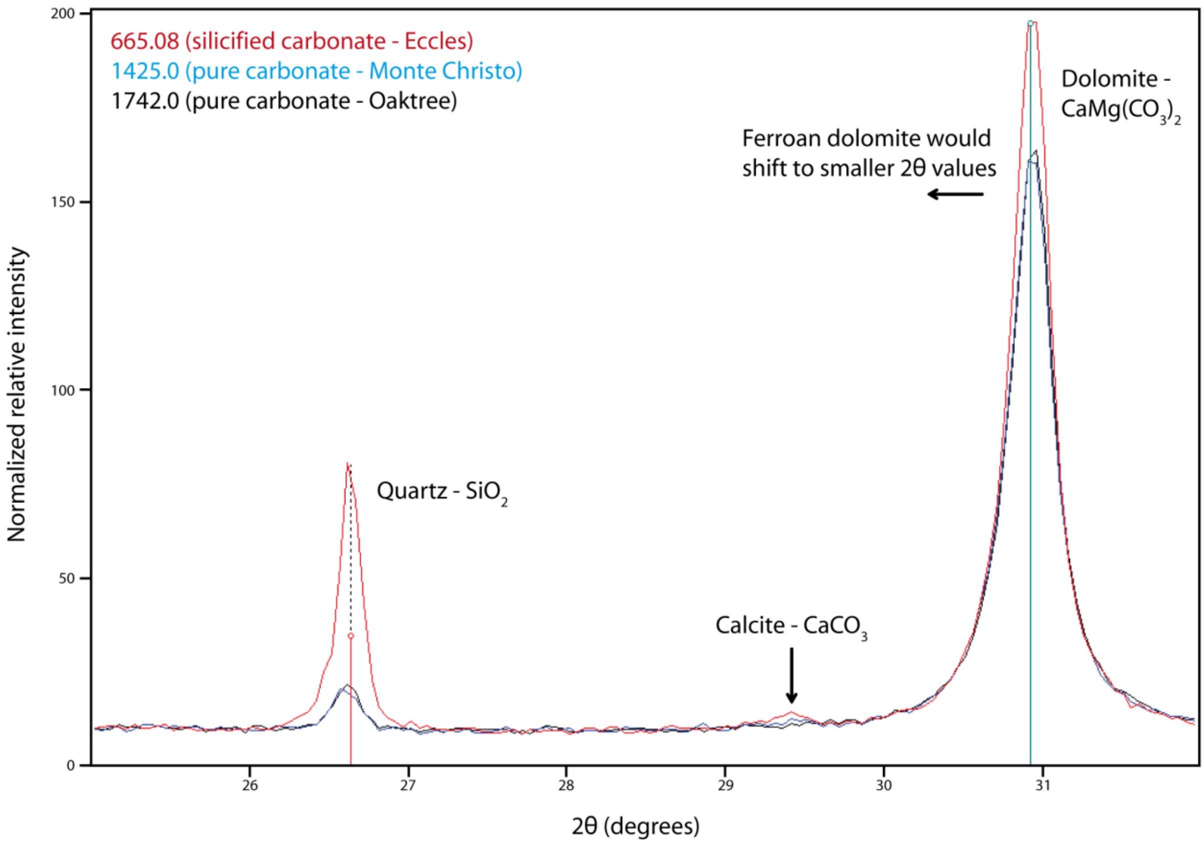

Fig. S3

XRD pattern of three representative carbonate samples from KMF-5 show a dolomite quartz mixture with minor amounts of calcite. No Fe minerals were detected with this method.

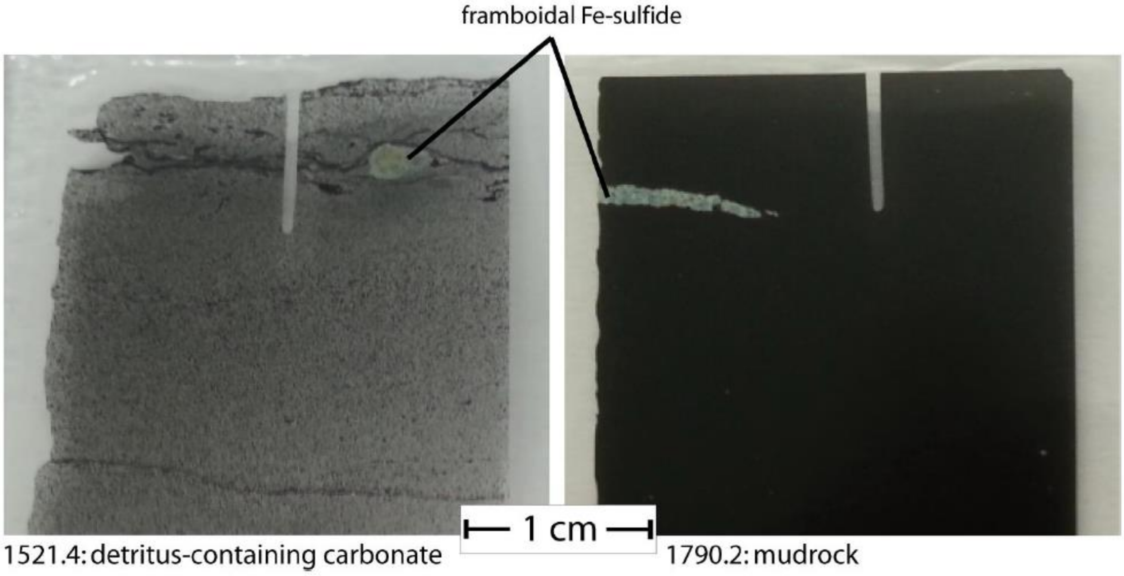

Fig. S4

Framboidal Fe-sulfide minerals in carbonate sample 1521.4 (Monte Christo Formation; KMF-5) and mudrock sample 1790.2 (Oaktree Formation; KMF-5) indicate oxygen-poor conditions with abundant organic carbon, iron and sulfur to form Fe-sulfides.

Additional Tables

Table SM-1: Sample-Standard-Bracketing Fe isotope results of single measurements of KMF-5 samples

Depth (m)	$\delta^{56}Fe_{avg}$ (‰)	$\delta^{57}Fe_{avg}$ (‰)	Analyses 1						Analyses 2						Analyses 3					
			$\delta^{56}Fe$ (‰)	2σ	$\delta^{57}Fe$ (‰)	2σ	$\delta^{58}Fe$ (‰)	2σ	$\delta^{56}Fe$ (‰)	2σ	$\delta^{57}Fe$ (‰)	2σ	$\delta^{58}Fe$ (‰)	2σ	$\delta^{56}Fe$ (‰)	2σ	$\delta^{57}Fe$ (‰)	2σ	$\delta^{58}Fe$ (‰)	2σ
665.08	-0.882	0.044	-0.851	0.040	-0.965	0.102	-1.353	0.592	-0.957	0.049	-1.457	0.090	-2.308	0.446	-0.840	0.044	-0.960	0.094	-1.376	0.663
665.18	-0.665	0.043	-0.646	0.045	-0.989	0.088	-1.576	0.478	-0.683	0.042	-1.042	0.082	-1.158	0.585						
673.84	-0.745	0.044	-0.753	0.046	-1.118	0.081	-1.958	0.560	-0.738	0.042	-1.119	0.085	-1.940	0.437						
674.55	-0.877	0.046	-0.897	0.049	-1.334	0.081	-1.938	0.491	-0.857	0.043	-1.273	0.084	-1.841	0.458						
678.60	-0.802	0.048	-0.807	0.044	-1.095	0.083	-1.011	0.486	-0.797	0.053	-1.217	0.075	-1.427	0.487						
680.58	-0.824	0.040	-0.834	0.032	-1.231	0.066	-1.344	0.441	-0.813	0.048	-1.264	0.092	-1.595	0.445						
682.70	-0.769	0.035	-0.731	0.034	-1.104	0.065	-1.219	0.371	-0.806	0.036	-1.181	0.066	-1.563	0.403						
703.30	-0.747	0.040	-0.724	0.040	-1.079	0.069	-1.595	0.478	-0.769	0.041	-1.134	0.076	-1.415	0.514						
711.80	-0.554	0.045	-0.562	0.047	-0.872	0.073	-1.036	0.495	-0.545	0.042	-0.827	0.078	-1.156	0.503						
875.50	-0.398	0.050	-0.415	0.052	-0.637	0.077	-1.465	0.522	-0.382	0.049	-0.637	0.085	-0.666	0.360						
884.83	-0.470	0.040	-0.489	0.036	-0.692	0.083	-0.508	0.455	-0.450	0.044	-0.720	0.088	-0.698	0.456						
921.78	-0.412	0.031	-0.397	0.034	-0.629	0.067	-0.706	0.438	-0.427	0.029	-0.580	0.070	-0.684	0.389						
1062.50	-0.672	0.034	-0.650	0.034	-0.925	0.065	-1.258	0.358	-0.695	0.034	-1.036	0.064	-1.175	0.347						
1072.73	-0.775	0.043	-0.772	0.042	-1.115	0.064	-1.409	0.362	-0.777	0.044	-1.158	0.069	-1.644	0.335						
1100.20	-0.658	0.035	-0.664	0.040	-0.954	0.064	-0.760	0.424	-0.652	0.031	-0.986	0.071	-1.390	0.296						
1136.75	-0.681	0.033	-0.654	0.029	-0.949	0.066	-1.288	0.425	-0.708	0.037	-1.024	0.082	-1.480	0.304						
1143.70	-0.707	0.046	-0.742	0.048	-1.060	0.082	-1.472	0.461	-0.715	0.048	-1.062	0.076	-1.367	0.484	-0.665	0.043	-0.995	0.079	-1.462	0.404
1199.50	-0.401	0.043	-0.416	0.047	-0.621	0.079	-1.111	0.501	-0.385	0.040	-0.643	0.087	-1.039	0.440						
1202.58	-0.511	0.048	-0.538	0.050	-0.771	0.071	-1.057	0.498	-0.484	0.045	-0.733	0.103	-0.777	0.485						
1239.98	-0.398	0.048	-0.428	0.048	-0.644	0.078	-0.804	0.565	-0.408	0.052	-0.563	0.087	2.123	0.443	-0.358	0.045	-0.601	0.083	-0.540	0.556
1265.10	-0.036	0.034	-0.044	0.038	-0.057	0.071	-0.192	0.364	-0.028	0.030	-0.008	0.068	-0.005	0.362						
1350.90	-0.366	0.043	-0.429	0.042	-0.571	0.084	-0.667	0.450	-0.400	0.045	-0.595	0.073	-1.154	0.463	-0.269	0.040	-0.399	0.074	-1.203	0.397
1401.00	-0.354	0.044	-0.361	0.047	-0.571	0.093	-0.994	0.542	-0.346	0.041	-0.562	0.071	-0.288	0.500						
1403.80	-0.348	0.035	-0.338	0.037	-0.539	0.057	-0.797	0.326	-0.358	0.032	-0.497	0.058	-0.722	0.449						
1406.80	-0.333	0.039	-0.298	0.038	-0.431	0.062	-0.239	0.317	-0.369	0.041	-0.568	0.079	-0.702	0.385						
1420.90	-0.463	0.048	-0.475	0.053	-0.762	0.076	-1.126	0.552	-0.452	0.042	-0.721	0.075	-0.711	0.547						
1425.40	-0.361	0.040	-0.335	0.035	-0.483	0.073	-0.771	0.311	-0.387	0.044	-0.530	0.062	-0.669	0.391						
1435.25	-0.350	0.047	-0.350	0.048	-0.594	0.092	-0.724	0.488	-0.350	0.046	-0.574	0.091	-0.556	0.464						
1442.17	-0.664	0.043	-0.656	0.050	-0.949	0.088	1.707	0.486	-0.699	0.040	-1.069	0.089	-1.419	0.615	-0.638	0.041	-1.058	0.077	-1.071	0.508
1454.61	-0.411	0.045	-0.430	0.044	-0.662	0.079	-1.003	0.565	-0.392	0.046	-0.698	0.082	-0.543	0.502						
1461.80	-0.407	0.036	-0.429	0.036	-0.635	0.068	-1.361	0.491	-0.386	0.036	-0.664	0.081	-0.808	0.503						
1464.30	-0.499	0.039	-0.519	0.041	-0.732	0.081	-1.045	0.504	-0.480	0.037	-0.767	0.067	-1.341	0.471						
1467.10	-0.372	0.038	-0.406	0.041	-0.662	0.075	-1.023	0.437	-0.337	0.036	-0.527	0.076	-1.381	0.530						
1475.35	-0.397	0.043	-0.435	0.041	-0.661	0.085	-1.373	0.493	-0.360	0.044	-0.596	0.071	-1.464	0.507						
1484.80	-0.280	0.037	-0.260	0.036	-0.388	0.067	-0.571	0.384	-0.299	0.039	-0.430	0.064	-0.504	0.363						
1491.85	-0.351	0.038	-0.381	0.040	-0.588	0.082	-0.727	0.471	-0.321	0.036	-0.533	0.073	-0.989	0.495						
1499.85	0.201	0.047	0.140	0.050	0.270	0.079	0.439	0.522	0.169	0.044	0.239	0.084	-0.144	0.460	0.295	0.046	0.443	0.088	-0.279	0.441
1524.70	-0.406	0.037	-0.418	0.039	-0.607	0.063	-0.946	0.372	-0.393	0.035	-0.532	0.066	-0.889	0.345						
1539.90	-0.378	0.041	-0.396	0.040	-0.564	0.079	-0.592	0.524	-0.436	0.042	-0.603	0.069	-1.222	0.855	-0.303	0.042	-0.464	0.088	-0.944	0.439

Table SM-1 continued

1551.70	-0.269	0.048	-0.299	0.049	-0.435	0.082	-0.770	0.435	-0.239	0.047	-0.370	0.074	-1.330	0.485						
1557.70	0.788	0.048	0.761	0.051	1.102	0.083	1.080	0.433	0.815	0.045	1.170	0.073	0.998	0.468						
1558.88	-0.253	0.040	-0.297	0.039	-0.405	0.085	-1.738	0.983	-0.262	0.034	-0.335	0.067	-0.837	0.541	-0.199	0.049	-0.286	0.088	-1.062	0.463
1574.15	-0.232	0.040	-0.280	0.038	-0.376	0.101	-0.521	0.432	-0.224	0.037	-0.341	0.063	-0.655	0.601	-0.192	0.045	-0.287	0.092	-0.698	0.430
1574.25	-0.204	0.051	-0.231	0.049	-0.336	0.093	-0.707	0.534	-0.177	0.054	-0.248	0.085	-0.763	0.470						
1574.30	-0.235	0.045	-0.291	0.051	-0.447	0.089	0.052	1.444	-0.230	0.047	-0.322	0.079	-3.534	0.437	-0.184	0.037	-0.257	0.081	-0.841	0.504
1589.75	-0.271	0.045	-0.308	0.045	-0.446	0.068	0.017	1.439	-0.203	0.043	-0.322	0.095	-0.748	0.426	-0.303	0.045	-0.443	0.080	-0.593	0.472
1589.90	-0.218	0.044	-0.251	0.050	-0.375	0.084	-0.541	0.448	-0.254	0.039	-0.402	0.068	0.050	0.417	-0.151	0.043	-0.234	0.092	-0.760	0.440
1604.60	-0.282	0.032	-0.243	0.031	-0.396	0.062	-0.406	0.465	-0.321	0.032	-0.501	0.073	-0.783	0.333						
1673.30	0.378	0.049	0.335	0.040	0.460	0.075	0.318	0.558	0.420	0.057	0.621	0.082	-0.285	0.470						
1731.10	-0.616	0.041	-0.631	0.040	-0.945	0.074	-1.109	0.452	-0.600	0.042	-0.851	0.063	-1.247	0.333						
1731.30	-0.668	0.029	-0.705	0.028	-1.008	0.066	-1.394	0.334	-0.631	0.031	-0.905	0.069	-1.040	0.352						
1742.30	-0.718	0.040	-0.710	0.041	-1.028	0.058	-1.684	0.422	-0.726	0.038	-1.041	0.070	-1.384	0.339						
1776.00	0.616	0.043	0.583	0.044	0.854	0.095	1.099	0.460	0.650	0.043	1.025	0.074	0.636	0.452						
1790.10	0.083	0.044	0.062	0.049	-0.011	0.081	0.134	0.481	0.103	0.039	0.107	0.088	-0.605	0.411						
1800.10	-0.318	0.042	-0.354	0.050	-0.552	0.070	-0.535	0.494	-0.282	0.035	-0.460	0.076	-1.324	0.433						
1800.30	0.439	0.045	0.397	0.049	0.590	0.075	-4.948	0.532	0.482	0.042	0.680	0.084	0.463	0.404						
1811.20	0.240	0.043	0.201	0.043	0.337	0.072	-0.318	0.524	0.279	0.043	0.390	0.077	-0.326	0.550						

2σ: 2 sigma standard deviation of 20 measurement cycles per sample analysis on ICP-MS

Table SM-2: Sample-Standard-Bracketing Fe isotope results of single measurements of BH-1 samples

Depth (m)	$\delta^{56}Fe_{avg}$ (‰)	$\delta^{57}Fe_{avg}$ (‰)	Analyses 1						Analyses 2						Analyses 3					
			$\delta^{56}Fe$ (‰)	2σ	$\delta^{57}Fe$ (‰)	2σ	$\delta^{58}Fe$ (‰)	2σ	$\delta^{56}Fe$ (‰)	2σ	$\delta^{57}Fe$ (‰)	2σ	$\delta^{58}Fe$ (‰)	2σ	$\delta^{56}Fe$ (‰)	2σ	$\delta^{57}Fe$ (‰)	2σ	$\delta^{58}Fe$ (‰)	2σ
340	-1.819	-2.650	-1.820	0.041	-2.635	0.072	-3.612	0.485	-1.817	0.038	-2.665	0.059	-3.294	0.497						
375	-0.854	-1.264	-0.846	0.039	-1.233	0.072	-1.474	0.544	-0.862	0.041	-1.294	0.067	-1.629	0.462						
488	-0.907	-1.282	-0.907	0.051	-1.282	0.091	-1.848	0.418												
670	-0.558	-0.814	-0.565	0.050	-0.837	0.069	-1.405	0.417	-0.550	0.036	-0.791	0.078	-1.202	0.423						
751	-0.926	-1.359	-0.944	0.037	-1.317	0.077	-1.851	0.446	-0.907	0.039	-1.401	0.077	-1.601	0.458						
1235	-0.721	-1.068	-0.730	0.041	-1.048	0.076	-1.515	0.383	-0.711	0.045	-1.087	0.075	-1.463	0.432						
1400	-0.897	-1.248	-0.831	0.049	-1.308	0.084	-1.827	0.426	-0.946	0.043	-1.212	0.079	-1.260	0.656	-0.914	0.055	-1.225	0.080	-1.591	0.491
1425	-0.803	-1.195	-0.814	0.036	-1.193	0.058	-1.898	0.484	-0.791	0.051	-1.197	0.071	-1.336	0.464						
1455	-0.804	-1.194	-0.818	0.036	-1.202	0.072	-1.809	0.437	-0.789	0.040	-1.186	0.074	-1.364	0.451						
1490	-0.498	-0.779	-0.495	0.044	-0.756	0.091	-1.058	0.498	-0.500	0.030	-0.801	0.084	-0.306	0.478						
1520	-0.604	-0.882	-0.612	0.041	-0.959	0.074	-1.369	0.375	-0.610	0.047	-0.843	0.081	-1.198	0.510	-0.591	0.043	-0.843	0.083	-1.124	0.421
1620	-0.604	-0.912	-0.594	0.044	-0.864	0.084	-0.906	0.498	-0.613	0.040	-0.959	0.080	-1.658	0.489						
1750	-1.207	-1.817	-1.219	0.045	-1.809	0.092	-2.469	0.411	-1.195	0.042	-1.824	0.073	-2.506	0.449						
1776	-0.721	-1.096	-0.742	0.048	-1.097	0.073	-1.422	0.406	-0.699	0.042	-1.094	0.076	-1.231	0.451						
1920	-0.951	-1.325	-1.077	0.054	-1.312	0.076	-1.351	0.789	-0.902	0.038	-1.313	0.073	-1.793	0.507	-0.873	0.041	-1.351	0.091	-1.547	0.467
2041	-0.790	-1.190	-0.800	0.030	-1.220	0.060	-1.590	0.340	-0.780	0.030	-1.160	0.070	-1.360	0.390						
2066	-0.570	-0.850	-0.603	0.043	-0.835	0.067	-1.368	0.482	-0.536	0.038	-0.865	0.081	-1.037	0.491						
2098	-0.225	-0.355	-0.230	0.030	-0.390	0.080	-0.490	0.370	-0.220	0.030	-0.320	0.070	-0.450	0.400						
2250	-1.065	-1.570	-1.080	0.040	-1.600	0.060	-2.060	0.380	-1.050	0.030	-1.540	0.060	-2.350	0.350						
2275	-1.037	-1.547	-1.047	0.045	-1.545	0.070	-1.857	0.465	-1.027	0.039	-1.549	0.073	-1.987	0.421						
2293	-1.080	-1.625	-1.110	0.040	-1.670	0.070	-2.290	0.390	-1.050	0.040	-1.580	0.080	-2.070	0.340						
2355	-1.204	-1.851	-1.215	0.034	-1.886	0.068	-2.303	0.462	-1.193	0.038	-1.815	0.073	-1.807	0.469						
2379	-1.068	-1.608	-1.075	0.033	-1.587	0.058	-2.287	0.442	-1.061	0.036	-1.629	0.085	-1.979	0.405						
2400	-1.235	-1.805	-1.250	0.030	-1.810	0.070	-2.500	0.400	-1.220	0.040	-1.800	0.080	-2.220	0.370						

2σ: 2 sigma standard deviation of 20 measurement cycles per sample analysis on ICP-MS

Table SM-3: Sample-Standard-Bracketing Fe isotope results of single measurements of Kuruman Kop outcrop samples

Sample Name	$\delta^{56}Fe_{avg}$ (‰)	$\delta^{57}Fe_{avg}$ (‰)	Analyses 1						Analyses 2					
			$\delta^{56}Fe$ (‰)	2σ	$\delta^{57}Fe$ (‰)	2σ	$\delta^{58}Fe$ (‰)	2σ	$\delta^{56}Fe$ (‰)	2σ	$\delta^{57}Fe$ (‰)	2σ	$\delta^{58}Fe$ (‰)	2σ
KU 12/04	0.453	0.645	0.413	0.042	0.581	0.086	0.911	0.515	0.493	0.042	0.709	0.091	0.610	0.438
KU 12/06	-1.744	-2.586	-1.765	0.053	-2.639	0.089	-4.116	0.507	-1.723	0.061	-2.533	0.095	-4.747	0.419
KU 12/26	-0.289	-0.421	-0.315	0.040	-0.466	0.081	2.464	0.567	-0.262	0.043	-0.375	0.087	-0.836	0.511
KU 12/25	-0.695	-1.015	-0.740	0.044	-1.055	0.072	-0.976	0.509	-0.650	0.048	-0.974	0.091	-1.660	0.489
KU 12/31	-0.955	-1.374	-1.010	0.038	-1.462	0.085	0.775	0.533	-0.899	0.044	-1.285	0.084	-2.501	0.451

2σ: 2 sigma standard deviation of 20 measurement cycles per sample analysis on ICP-MS

Table SM-4: Fe mineral standards used for linear component analyses

Mineral	Structural formula	Source	Reference
Ankerite	$Ca(Fe(II),Mg, Mn)(CO_3)_2$	natural	E. Swanner
Siderite	$Fe(II)(CO_3)$	synthetic	T. Borch, C. Hansel, S. Fendorf
Goethite	α-Fe(III)O(OH)	natural	Sirine Fakra, Matthew A. Marcus
Magnetite	Fe_3O_4	natural	J. Frommer and A. Voegelin
Pyrite	FeS_2	natural	J. Frommer and A. Voegelin
Marcasite	FeS_2		E. Swanner
Chlorite (Ripidolite - CCa2)	$Ca_{0.5}(Mg_{4.44},Fe(III)_{3.47},Fe(II)_{3.02},Al_{0.60},Mn_{0.01},Ti_{0.06})(Si_{4.51},Al_{3.49})O_{20}(OH)_{16}$	Clay Mineral Society	Source Clay Repository (2001) Source clay physical/chemical data: http://web.missouri.edu/~geoscjy/SourceClay/chem.html. The Clay Minerals Society
Ferrosmectite (Fe-bearing montmorillonite)	$(Na_{0.48}Ca_{0.03}K_{0.01})(Al_{1.54}Mg_{0.33}Fe(III)_{0.09}Fe(II)_{0.02})(Si_{3.87}Al_{0.13})\ O_{10}(OH)_2\cdot nH_2O$	natural	T. Borch, C. Hansel, S. Fendorf, J.W. Stucki

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