

SILTSTONES ACROSS THE *DAPTOCEPHALUS* (*DICYNODON*) AND  
*LYSTROSAURUS* ASSEMBLAGE ZONES, KAROO BASIN, SOUTH AFRICA, SHOW  
NO EVIDENCE FOR ARIDIFICATION

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Supplemental Information

METHODS

**Elemental composition by XRF.**— Elemental composition of areas displaying a homogenous matrix in thin section and the clay fractions of 23 hand samples were obtained using a Bruker M4 Tornado Micro-XRF spectrometer, and the data were analyzed with the M4 Tornado analytic software (ver. 1.2.0.2687; Supplemental Data Table 1). For each thin section, three to eight field-of-view-sized regions were marked on the slide under 100X magnification, with more regions selected for those samples displaying greater lithologic heterogeneity where greater geochemical variation was likely. Data were acquired from a minimum of 233 and a maximum of 649 sampling points per thin section, manually chosen in the Micro-XRF under 100X magnification. The mud fraction of 21 samples (marked b and c in Supplemental Data Table 1) was separated after any surficial iron oxyhydroxide staining on weathered samples was removed physically, and then by powdering the sample with a mortar and pestle. These samples were hand ground to powders using a mortar and pestle and made into clay slides using the modified Drever's filter-membrane peel technique by Pollastro (1982). Data were acquired from a minimum of 48 and a maximum of 123 sampling points per slide, manually chosen in the Micro-XRF under 10X magnification. All samples were run for the region between 0.25 keV and 20.00 keV, with a live time of 30 s

per sampling point (30  $\mu\text{m}$ ), and the following elements were quantified: Si, Al, K, Fe, Na, Mg, Ca, Ti, Mn, P, and S.

**Clay mineralogy by XRD.**— The clay mineralogy of 19 samples (marked c in Supplemental Data Table 1) was determined at Southern Methodist University, Dallas, Texas, in Neil Tabor's laboratory. The clay mineralogy of these 19 untreated mud slides are referred to as the bulk slides, hereafter. Powders were suspended in centrifuge tubes filled with deionized water ( $\text{dH}_2\text{O}$ ) and deflocculated in a Chicago Electric Power Tools Industrial 91957 ultrasonic cleaner for ~10 minutes. Then, the samples were placed in an Eppendorf 5810 centrifuge for 3 minutes at 750 rpm to separate the silt-and-clay fractions, and the supernatant of each used to make clay slides following Pollastro (1982). Three treatments that allow for the identification of specific clay minerals—KCl,  $\text{MgCl}_2$ , and Glycerol—were applied to the clay films before being transferred onto petrographic slides. Nineteen samples were treated in this way for a total of 57 prepared slides. A fourth heat treatment, used to help distinguish between kaolinite and chlorite, was applied to KCl-treated slides after they were run in the XRD for data collection. These slides were baked at  $550^\circ\text{C}$  for 3 hours. All slides were scanned for the  $2\theta$  range of  $2^\circ$  to  $30^\circ$ , with a scan speed of  $1^\circ 2\theta / \text{min}$  and a step width of  $0.01^\circ 2\theta$  in a Rigaku Ultima III X-ray diffractometer in the Bragg-Brentano focusing beam configuration with a Cu X-ray source. Analysis of XRD data was conducted with the Rigaku PDXL2 software.

**$\text{Fe}^{2+}/\text{Fe}^{3+}$  Concentrations by Mössbauer Spectroscopy.**— Four samples (marked d in Supplemental Data Table 1) from section 10 were powdered, sieved, and sent to Darby Dyer's laboratory at Mount Holyoke College for Mössbauer spectroscopic analysis. These

were chosen on the basis of sample color, grain size, sedimentary structures, and stratigraphic position (Supplemental Data Table 1). Samples JL109-10 and JL109-3B are from the base of the section. Sample JL109-10 is mostly greenish-gray (5Y 4/1) with a thin reddish-gray interval at the bottom, and was collected ~ 1 m below JL109-3B, whereas JL109-3B is reddish-gray (5YR 3/1) in color (Supplemental Data Table 1; Fig. 3). Samples JL109-16 and JL109-18 come from the same interval at the top of the section 10 (Supplemental Data Table 1; Fig. 3). JL109-16 also is greenish-gray (5Y 4/1) in color but has a thin reddish-gray interval at the top (Supplemental Data Table 1; Fig. 7E), whereas JL109-18 is reddish-gray (5YR 3/1; Supplemental Data Table 1). Only the greenish-gray areas of JL109-16 and JL109-18 were powdered and submitted for Mössbauer spectroscopy. Approximately 40 mg of the sample were gently mixed with sugar, then heaped in a sample holder confined by kapton tape. Mössbauer spectra were acquired at 295K using a source of 90 mCi  $^{57}\text{Co}$  in Rh on a SEE Co. (formerly WEB Research Co.) model WT302 spectrometer (Mount Holyoke College). For each sample, the fraction of the baseline due to the Compton scattering of 122 keV gammas by electrons inside the detector was determined by measuring the count rate with and without a 14.4-keV stop filter (~2 mm of Al foil) in the gamma beam. Compton-corrected absorption was calculated for each individual spectrum using the formulation  $A/(1 - b)$ , where  $b$  is the Compton fraction and  $A$  is the uncorrected absorption. This correction does not change the results of the fits per se but does allow accurate determination of % absorption in the spectra. It is necessary because the range of energy deposited in the detector by Compton events extends from 0 keV to 40 keV, overlapping both the 14 keV and 2 keV energies deposited by the 14 keV gammas.

Run times were 24-48 hours. Spectra were collected in 2048 channels and corrected for nonlinearity via interpolation to a linear velocity scale, which is defined by the spectrum of the 25  $\mu\text{m}$  Fe foil used for calibration. The WMOSS algorithm fits a straight line to the points defined by the published values of the Fe metal peak positions (as y values) and the observed positions in channels (x values). Data were then folded before fitting, using the WMOSS Auto-fold procedure that folds the spectrum about the channel value that produces the minimum least squares sum difference between the first half of the spectrum and the reflected second half of the spectrum.

Mössbauer spectra were modeled using the Mex\_disdd program, which was acquired from the University of Ghent courtesy of E. DeGrave. The program uses Lorentzian line shapes and solves full hyperfine interaction Hamiltonians for isomer shift and quadrupole splitting but uses multiple distributions for magnetically split spectral components, where the value of the magnetic field is given as the value for the most heavily weighted distribution. Errors on isomer shift, quadrupole splitting, and line widths are  $\pm 0.02$ - $0.03$  mm/s for the doublets and sextets in these spectra, and errors on hyperfine fields are  $\pm 2$ - $3$  kOe. Errors on total %Fe<sup>3+</sup> are  $\pm 1$ - $5$  % absolute based on repeated fits to the same spectra, with a detection limit for Fe<sup>3+</sup> of roughly 1%.

**Thin Section SEM Imaging.**— SEM imaging was undertaken at the University of Maine-Orono with the assistance of Martin Yates. Samples confirmed to have hematite by Mössbauer spectroscopy were scanned using a Tescan Vega II XMU tungsten filament SEM with backscatter capabilities to determine the distribution and morphology of metal-rich oxides and silicates. Thin sections were carbon-coated before scanning, and

mineralogy was determined based on semi-quantitative elemental composition data obtained by the EDAX Pegasus EDS system. Digital images and EDS X-ray point spectra of representative grains were collected.

#### LITERATURE CITED

POLLASTRO, R.M., 1982, A recommended Procedure for the preparation of oriented clay-mineral specimens for X-ray diffraction analysis: modifications to Drever's filter-membrane peel technique: United States Department of the Interior Geological Survey, Open-File Report 82-71.