

APPENDIX 1: DETAILED METHODS

Framboids from the Cariaco Basin and Demerara Rise were prepared into lamellar foils for STEM imaging analysis using a dual-beam scanning electron microscope (SEM; Thermo Fisher Scientific Helios Nanolab 600i) with focused-ion beam capabilities (FIB), at the Environmental Molecular Sciences Laboratory (EMSL) at Pacific National Northwest Laboratory (PNNL). Framboids of interest were identified with SEM and conventional FIB techniques were used for the extraction and preparation of the framboids. The approach is summarized for the Cariaco Basin specimen here, with the same approach applied to the Demerara Rise specimens. First, a protective Pt-C capping layer was deposited as a rectangular section of the identified framboid surface (e.g., $\sim 4 \times \sim 15 \mu\text{m}$) by electron beam induced deposition ($\sim 150 \text{ nm}$) followed by ion-beam induced deposition ($\sim 350 \text{ nm}$) using the dual-beam FIB microscope, further protecting the framboids from Ga^+ ion damage and Ga contamination during ion milling. The framboid substructure was viewed in cross-section by trenching both sides of the rectangular region of interest to a depth $\sim 10 \mu\text{m}$ by ion beam milling, using an 30kV accelerating voltage and $\sim 3 \text{ nA}$ current. After substructures of interest were identified, the section was extracted with an OmniProbe micromanipulator and transferred to a Cu TEM $\frac{1}{2}$ grid and thinned to $< 100 \text{ nm}$. The same preparation approach was applied to the Demerara Rise specimen.