

Supplementary Information

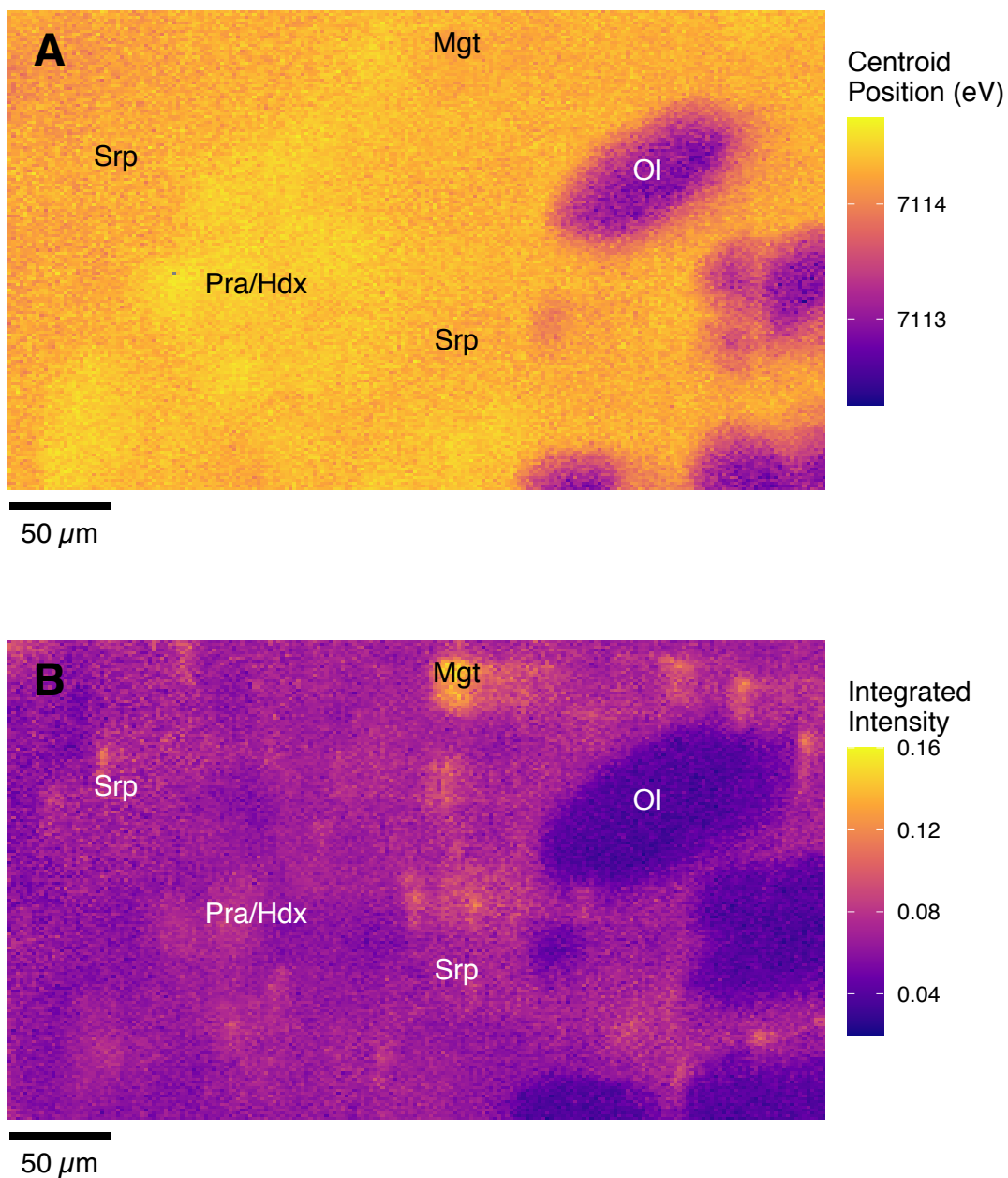
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Data and Analysis Scripts

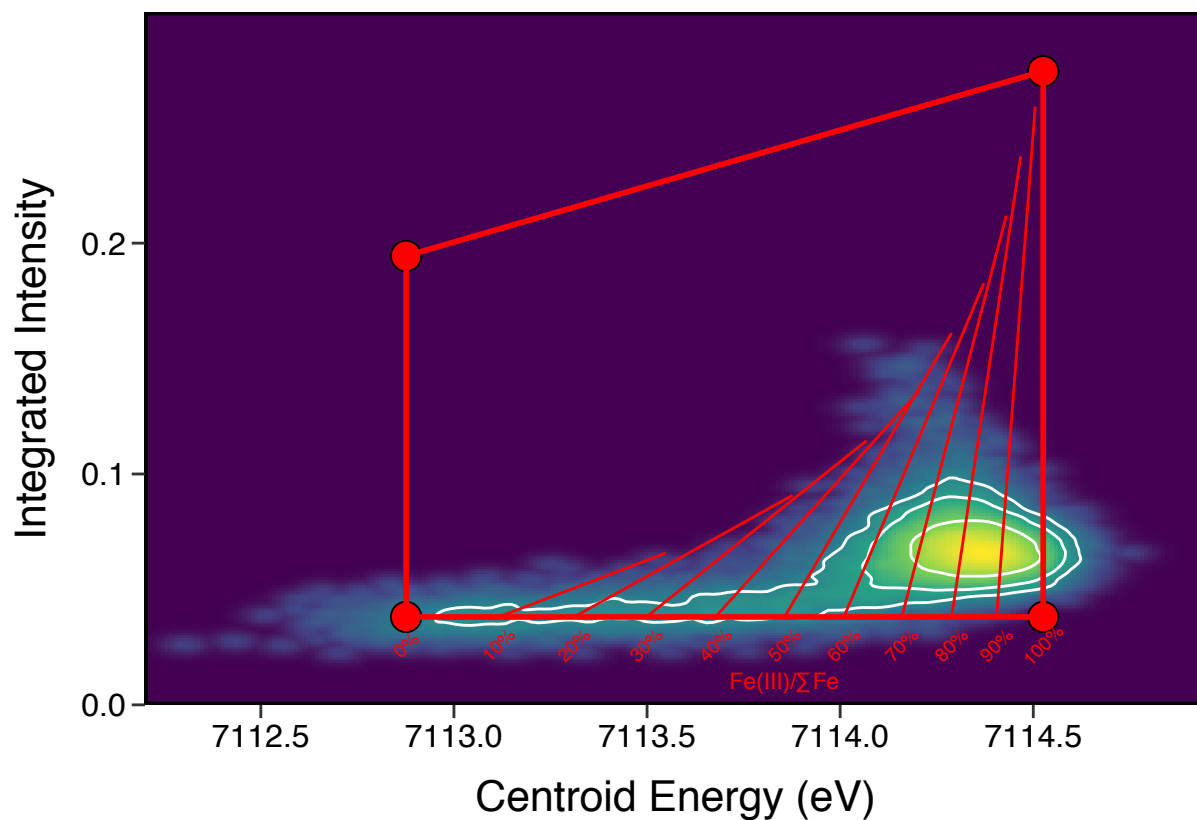
The raw X-ray data and all of the R code used for the processing and analysis of the maps, error analyses, and generation of figures, are available in a Github repository associated with this work. The data and code can be installed in R in order to replicate the analyses performed here. The code is available at:

<https://github.com/ellison2811/FeRedoxImaging>

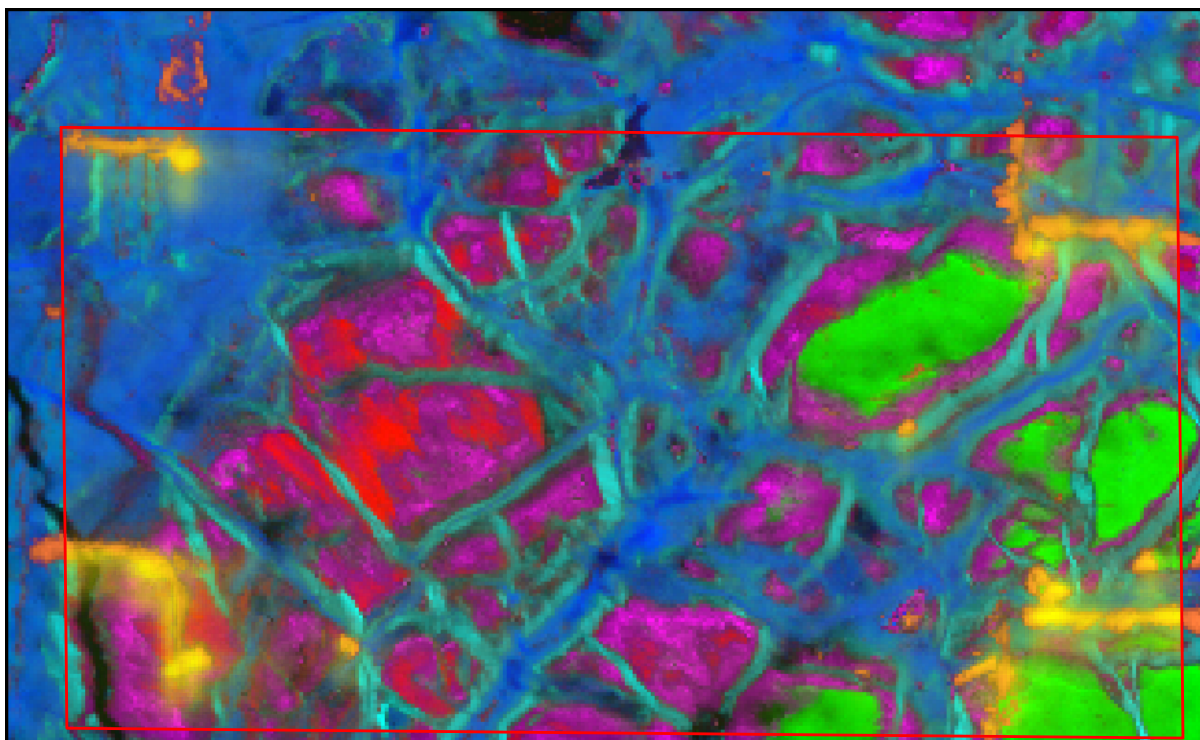
Supplemental Figures



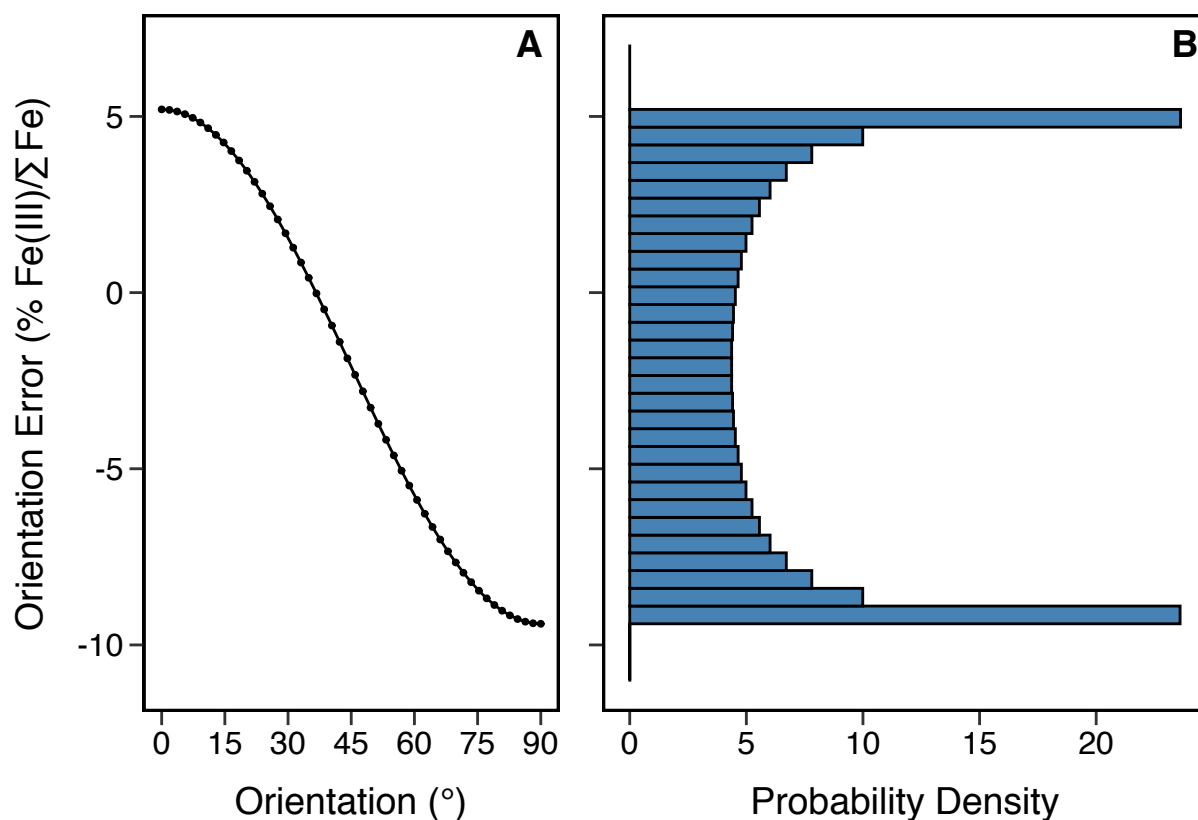
Supplemental Figure 1: Maps of pre-edge centroid position (A) and integrated intensity (B) prior to gaussian smoothing step. Abbreviations: Ol = olivine, Srp = Serpentine, Pra = Pyroaurite, Hdx = Hydroxide phase, Mgt = Magnetite.



Supplemental Figure 2: Fe redox variogram, showing kernel density of pixels without gaussian smoothing applied to the centroid and intensity maps. $\text{Fe(III)}/\Sigma\text{Fe}$ is given by the variogram tielines at 10% intervals. Contour line envelopes that encompass 50%, 75%, and 90% of the total pixel kernel density are also shown.



Supplemental Figure 3: Corregistration of the XRF and Raman data sets. Control points in the TiO_2 component of the Raman mineralogy map (orange) and the Ti X-ray fluorescence map were chosen in order to calculate and apply an affine transformation. The resulting transformation of the Ti X-ray fluorescence map is shown (yellow overlay with transparency).



Supplemental Figure 4: Distribution of errors due to orientation effects. Sine function fit to measured orientation errors in antigorite, which is given by $f(\varphi) = -0.021 + 0.073 \sin\left(\frac{2\pi}{180^\circ}\varphi + \frac{\pi}{2}\right)$, where φ is the orientation angle in degrees (A, modified from Muñoz et al. 2013). Arcsine probability density distribution resulting from the orientation error, which is given by $p(f) = \frac{1}{\pi\sqrt{0.073^2 - (-0.021 - f)^2}}$ for $-9.4\% < f < 5.2\%$ and $p(f) = 0$ everywhere else (B).

References

Muñoz, M., Vidal, O., Marcaillou, C., Pascarelli, S., Mathon, O., and Farges, F. (2013) Iron oxidation state in phyllosilicate single crystals using Fe-K pre-edge and XANES spectroscopy: Effects of the linear polarization of the synchrotron X-ray beam. *American Mineralogist*, 98, 1187–1197.