Electronic Supplementary Material (ESI) for Environmental Science: Processes & Impacts.

Supplementary data

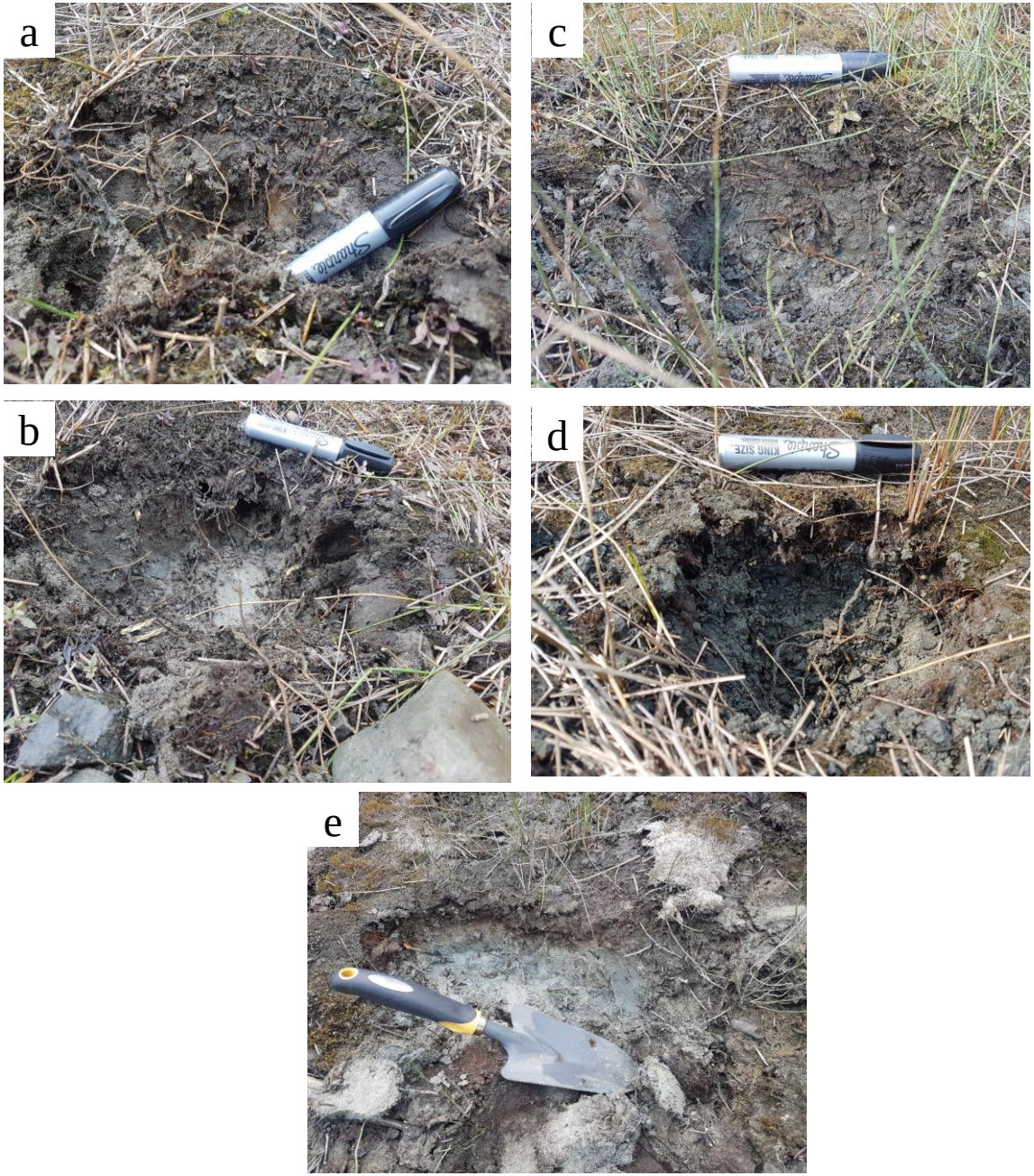

Figure S1: Photos of the samples and sample locations a) 1s, b) 2s and 2u, c) 3s and 3u, d) 4s and 4u, e) 5s and 5u

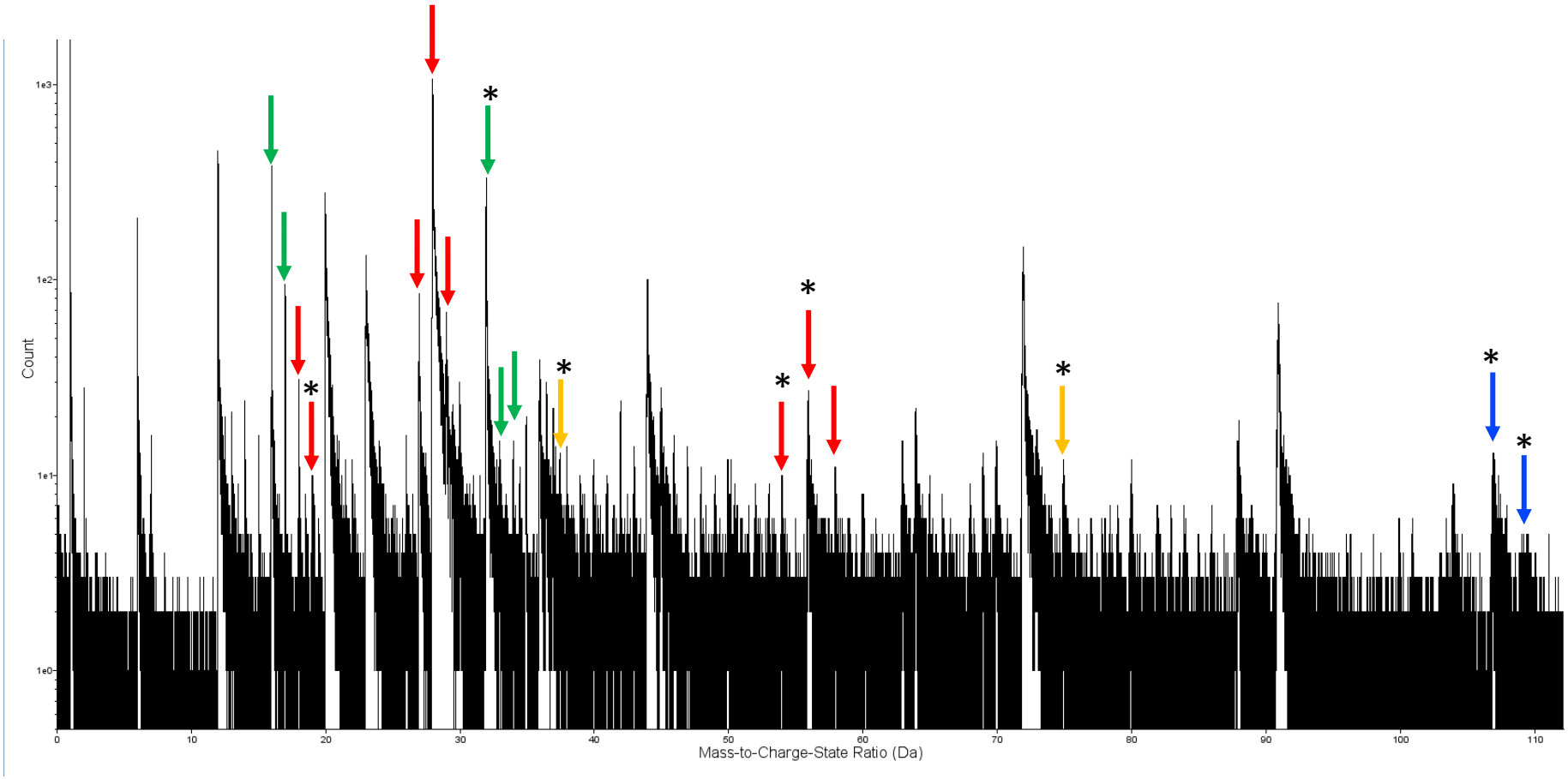

Figure S2: The mass spectrum of an APT-FIB section. The peaks for Fe, S, As, and Ag are indicated with red, green, orange, and blue color arrows, respectively. Peaks corresponding to only one of the above-mentioned elements are labelled with stars.

Table S1: contact pH values of the collected samples

Sample name	measurement with 0.01 mol L^{-1} $CaCl_2$ solution	measurement with distilled water
1s	6.52	7.21
2s	6.31	6.71
3s	6.30	6.86
4s	6.63	6.98
5s	6.16	6.84
2u	6.26	7.52
3u	5.89	6.25
4u	5.95	7.05
5u	6.45	7.29

Table S2: Concentrations of selected elements in the collected soil and tailings samples

Sample	Ag (mg kg-1)	As (mg kg-1)	Co (mg kg-1)	S (mg kg-1)	Fe (%)	Cu (mg kg-1)	Zn (mg kg-1)	Ni (mg kg-1)	Sulfide (%)	Organic C (%)
1s	89.4	944	688	1300	3.73	236	533	237	0.07	2.14
2s	129	726	437	1000	4.4	262	349	234	0.03	2.3
3s	20.5	3130	1210	3600	3.59	266	646	484	0.12	12.1
4s	124.7	12243	5240	12600	5.37	1695	575	9670	0.32	5.19
5s	18.5	2640	5810	5600	1.8	205	970	3020	0.14	12.4
2u	93.6	414	169	400	4.25	194	132	112	-	-
3u	32.3	376	150	900	6.38	391	199	119	-	-
4u	96.8	3110	1075	1300	6.02	524	331	801	-	-
5u	57.2	432	452	200	4.35	71	74	237	-	-

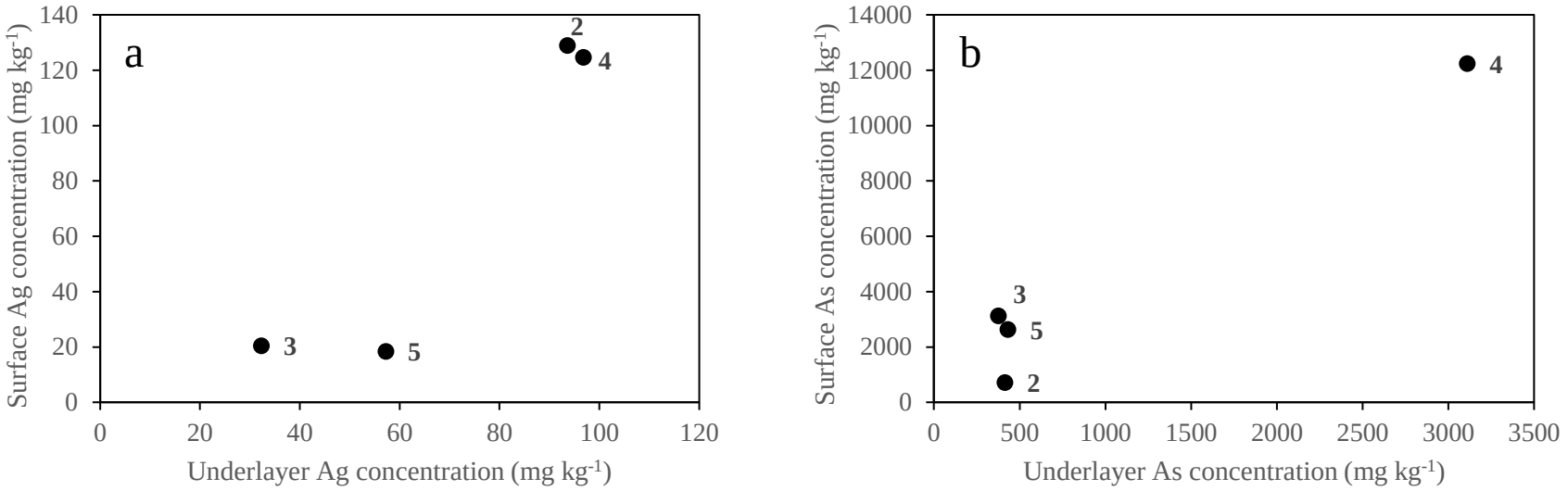

Figure S3: Plots of concentrations in the underlying tailings versus concentrations in organic-rich surficial soils: (a) Ag and (b) As.

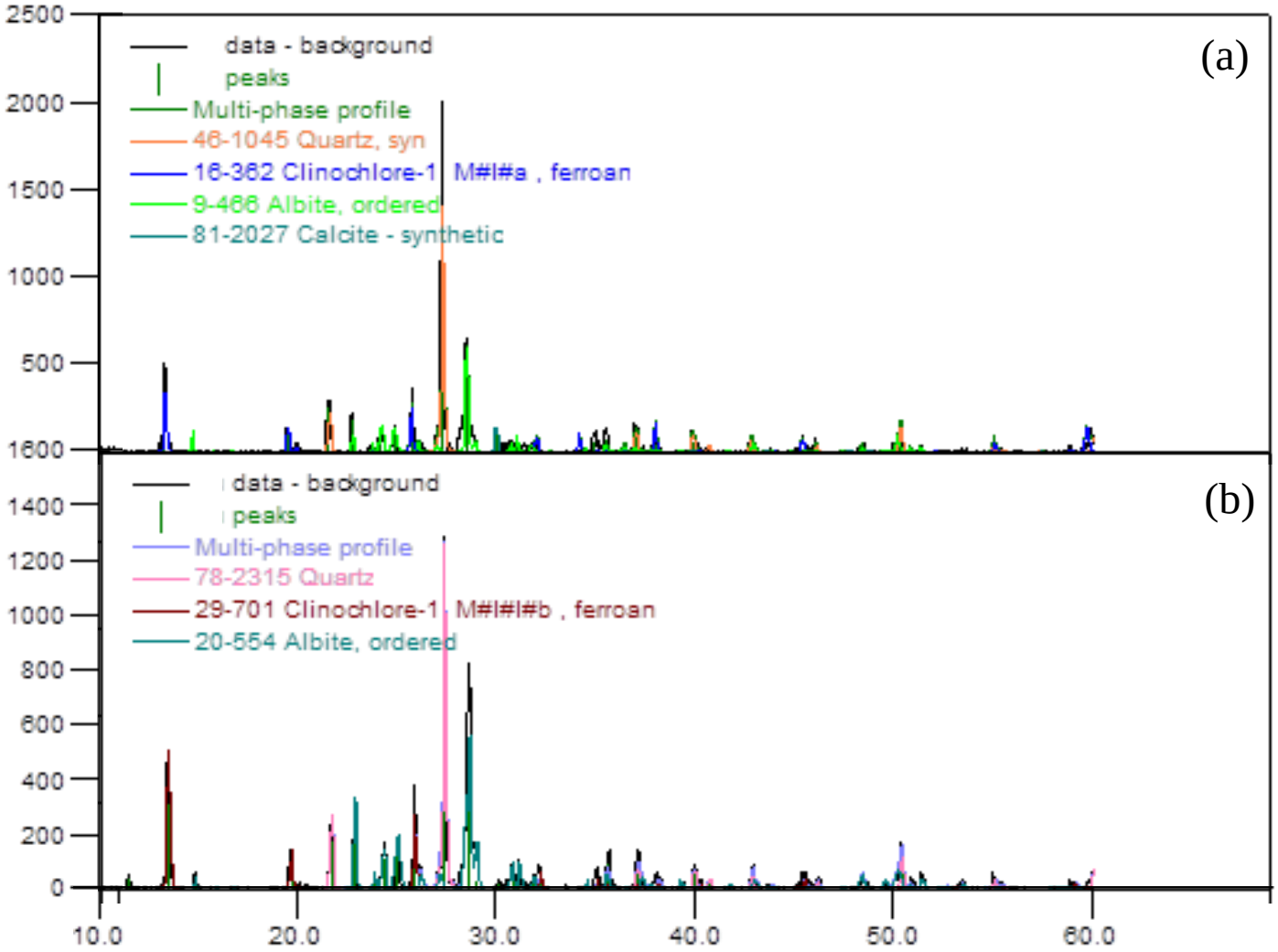

Figure S4: The XRD patterns of all collected samples were very similar, Here are shown the XRD patterns for the samples (a) 4s and (b) 4u

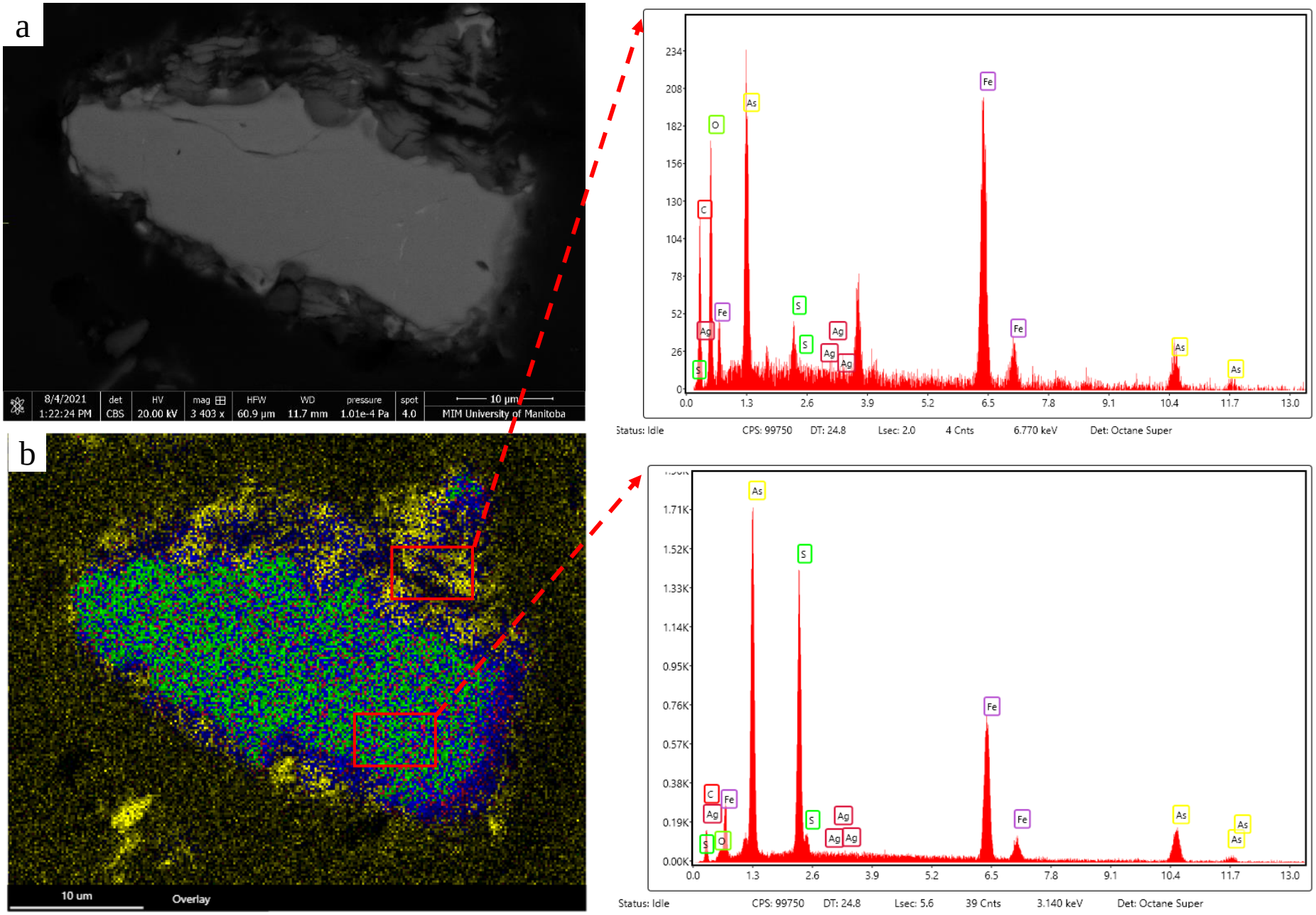

Figure S5: Arsenopyrite with scorodite-bearing mineral surface coating: (a) SEM image in BSE mode; (b) SEM-EDS chemical distribution map (S: green, Fe: blue, As: red, O: yellow) and EDS spectra for two selected locations

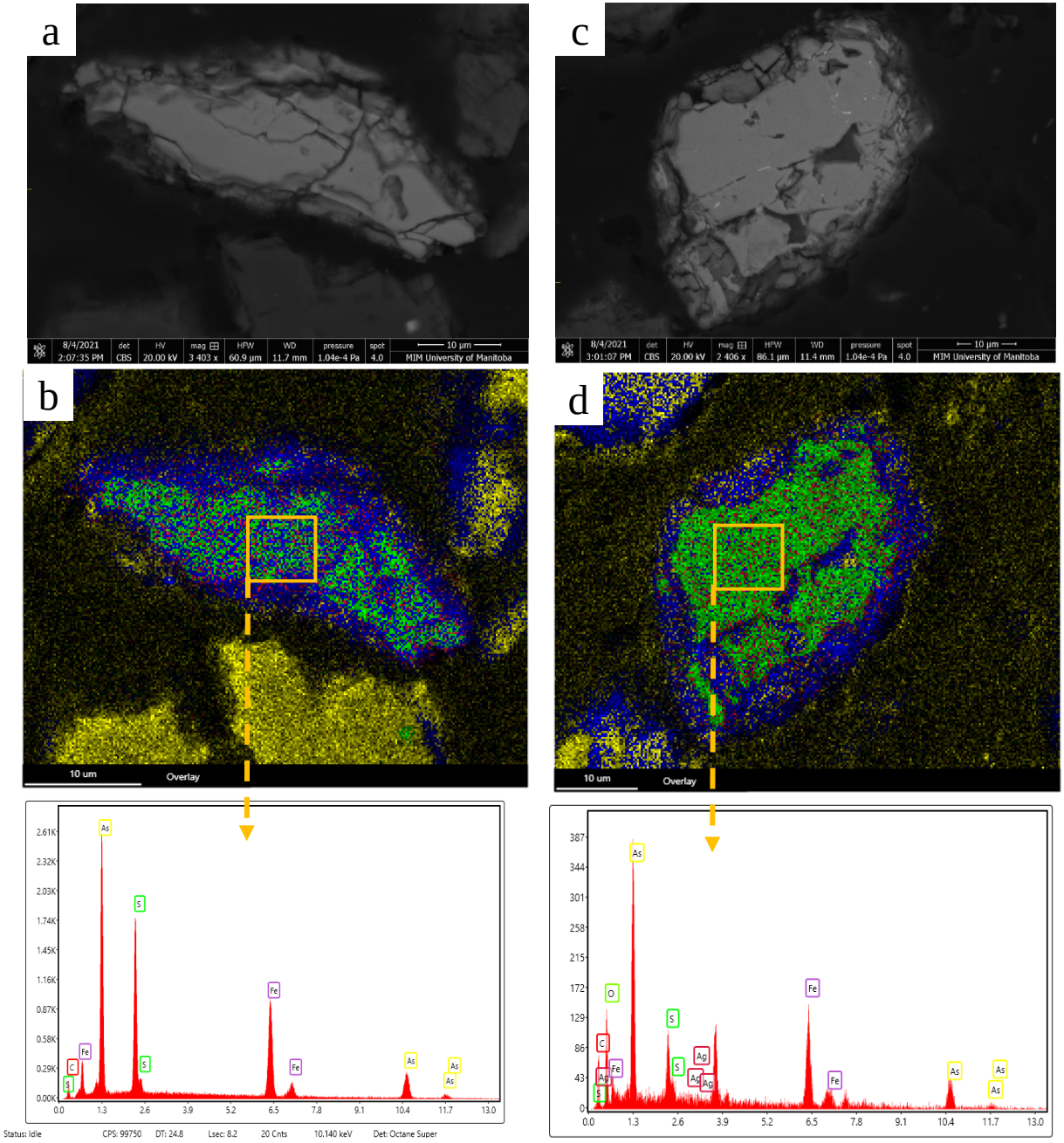

Figure S6: Arsenopyrite with scorodite-bearing mineral surface coating: (a, c) SEM images in BSE mode, (b, d) SEM-EDS chemical distribution maps (S: green, Fe: blue, As: red, O: yellow) and EDS spectra for two selected locations.

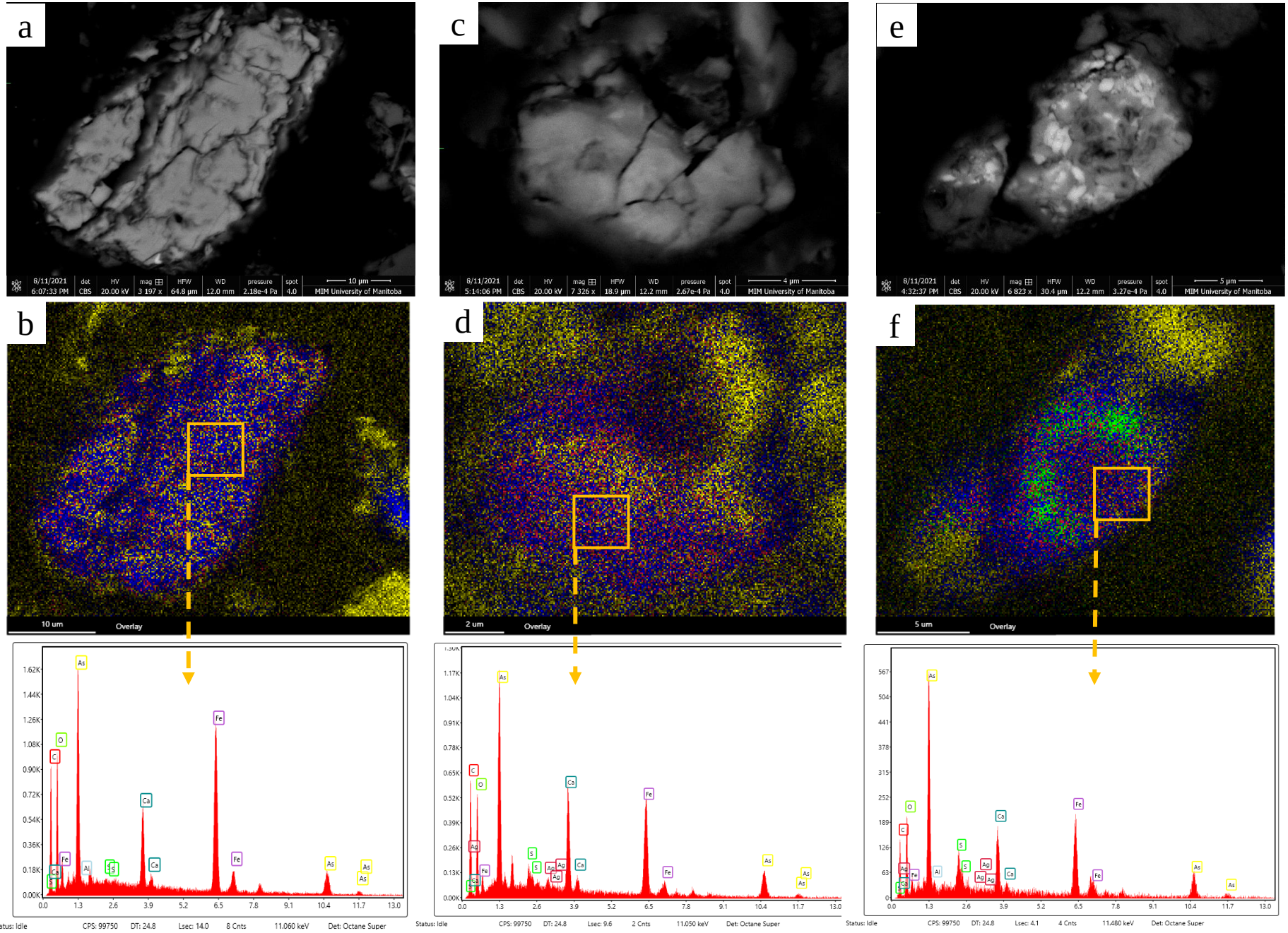

Figure S7: Scorodite-bearing mineral surface coatings on calcite: (a, c, e) SEM images in BSE mode and (b, d, f) SEM-EDS chemical maps with corresponding EDS spectra (S: green, Fe: blue, As: red, O: yellow)

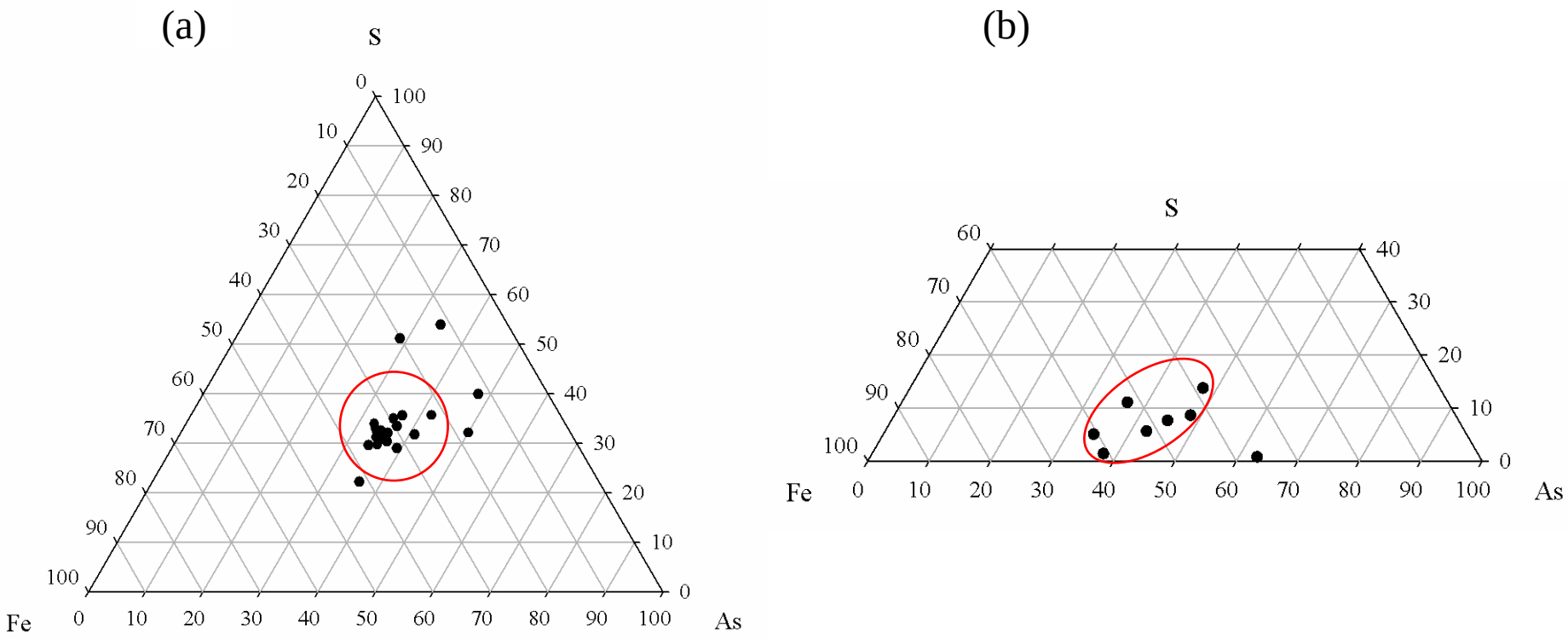

Figure S8: Triangle diagrams with Fe vs. As vs. S depicting the chemical composition of (a) sulfarsenides and (b) Sulfur-bearing arsenate

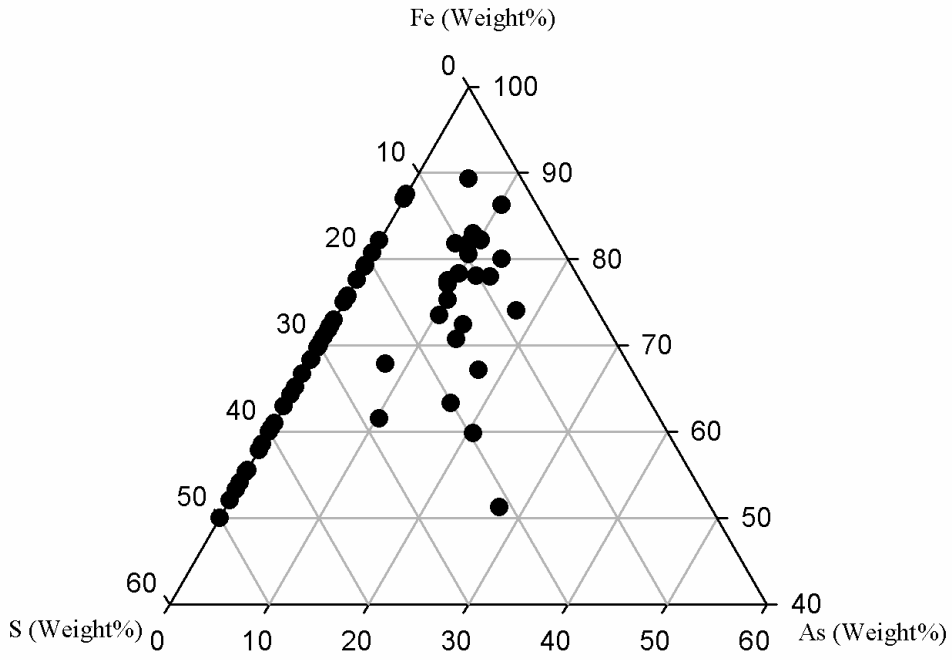

Figure S9: Triangle diagram depicting the Fe, S, and As content in 61 mineralized organic grains from four different sample locations

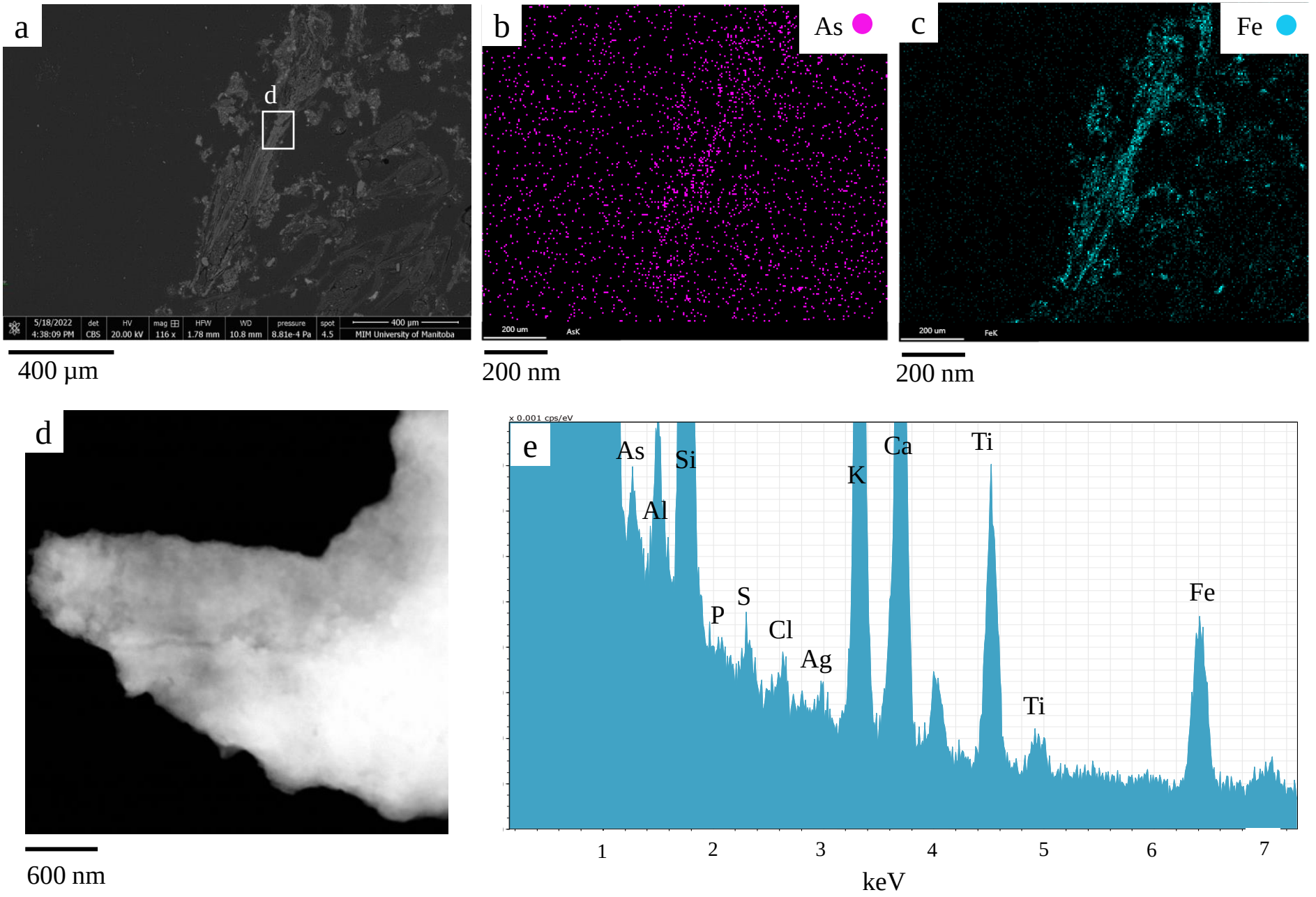

Figure S10: Images for TEM-mill 2: (a) SEM images in backscattering mode of an OM grain, the location of the extracted part for TEM analyses is shown as a rectangular; (b)-(c) SEM-EDS chemical distribution maps for (b) As (purple) and (c) Fe (blue) of the selected grains; (d) STEM image and (e) corresponding STEM-EDS spectrum for a selected area in the prepared TEM-mill1

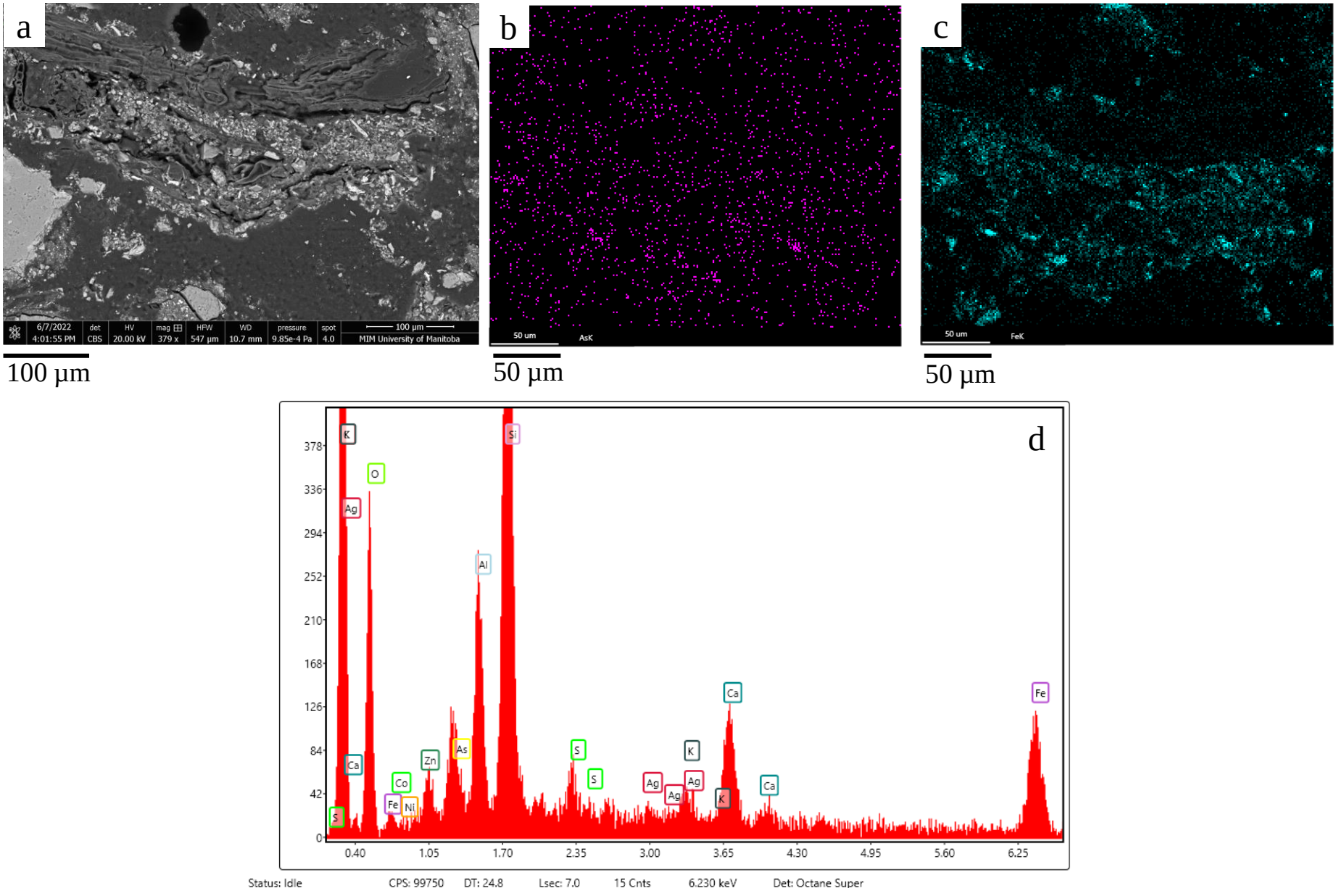

Figure S11: Images of an Ag-bearing OM grain: (a) SEM image in backscattering mode of an Ag-bearing OM grain; (b)-(c) SEM-EDS chemical distribution maps for (b) As (purple) and (c) Fe (blue) of the OM grain; (d) SEM-EDS spectrum of the entire OM grain.

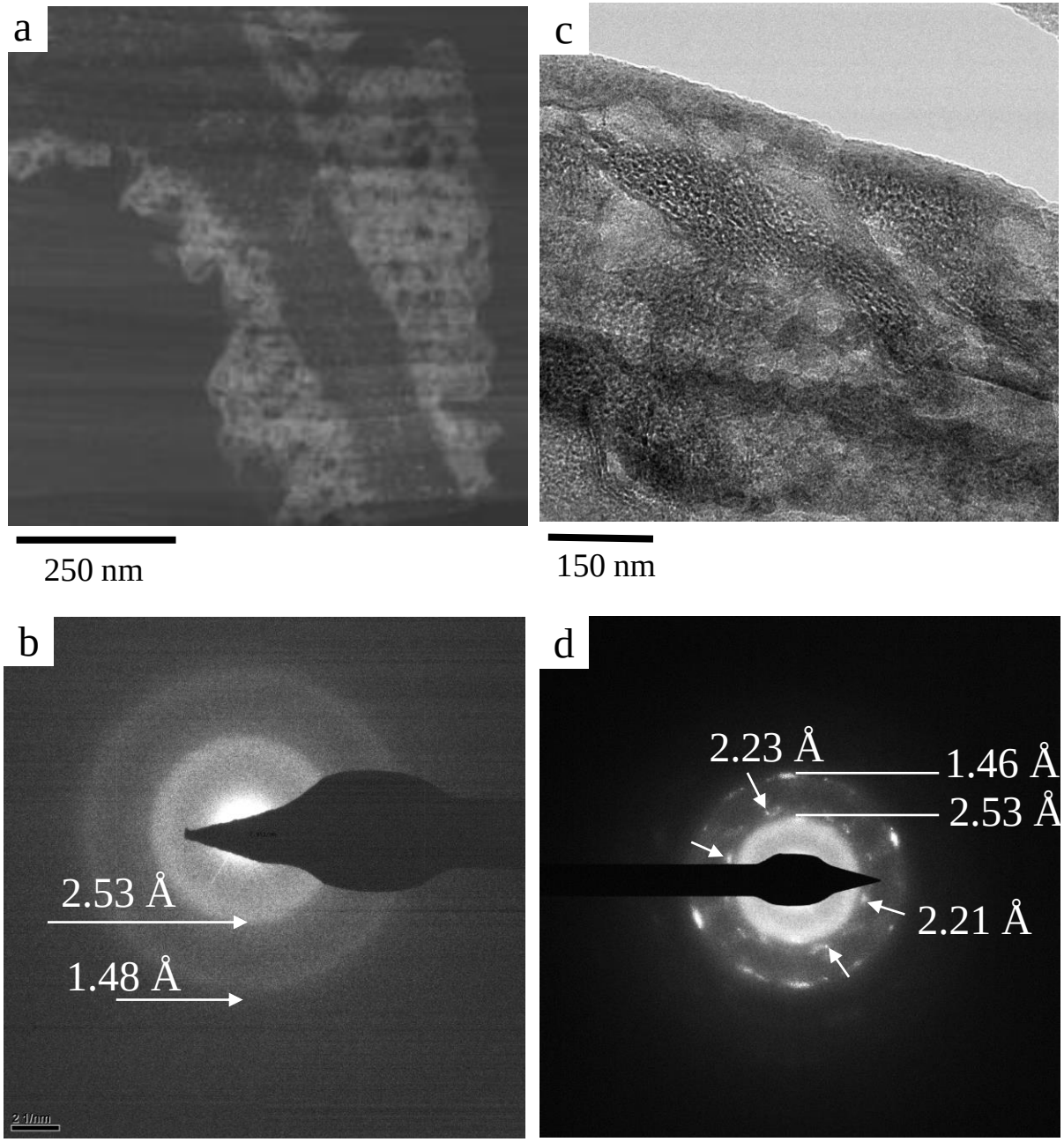

Figure S12: (a) STEM-EDS image of ferrihydrite with (b) corresponding diffraction pattern, (c) STEM-EDS image of hematite with (d) corresponding diffraction pattern,

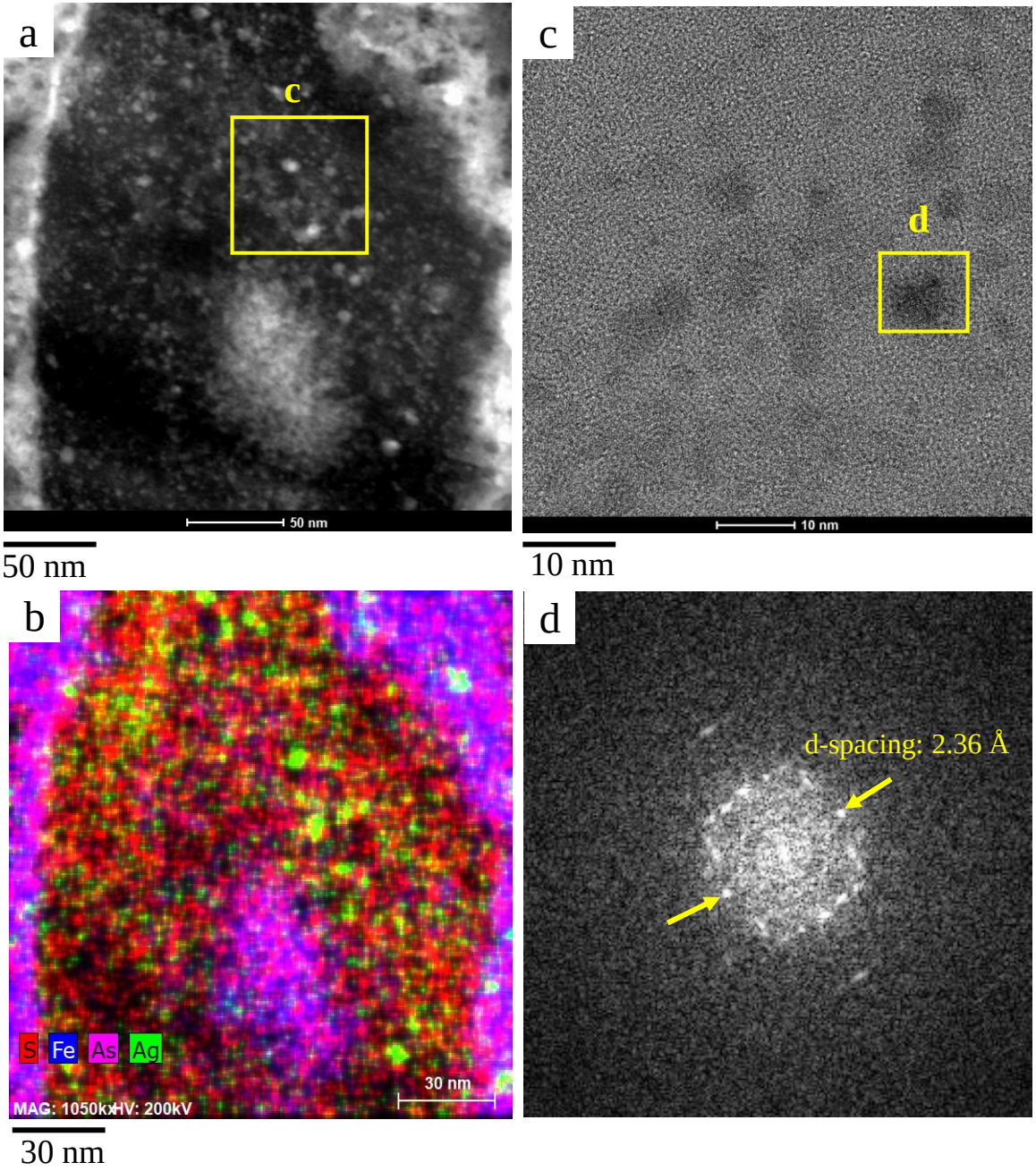

Figure S13: (a) STEM-EDS image and (b) STEM-EDS chemical distribution map (Fe: blue, As (violet), Ag (green) of an area with Ag NPs; (c) HRTEM image of an assemblage of Ag NPs and (d) FFT pattern of a selected Ag NP (labelled in (c)).

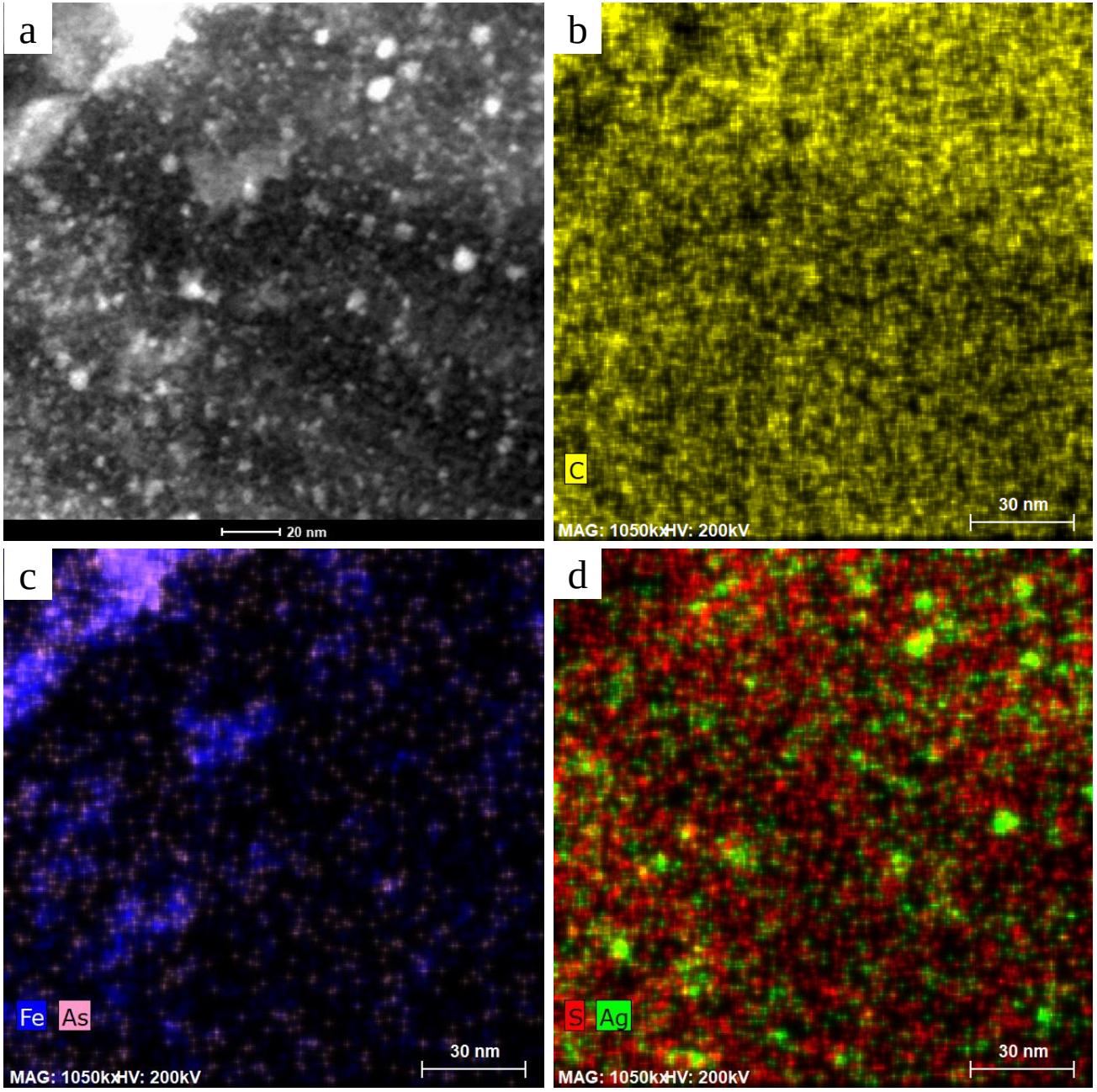

Figure S14: Occurrence of Ag and Ag_2S NP: (a) HRTEM image and (b)-(d) STEM-EDS chemical distribution map for (b) C, (c) Fe and S, and (d) S and Ag

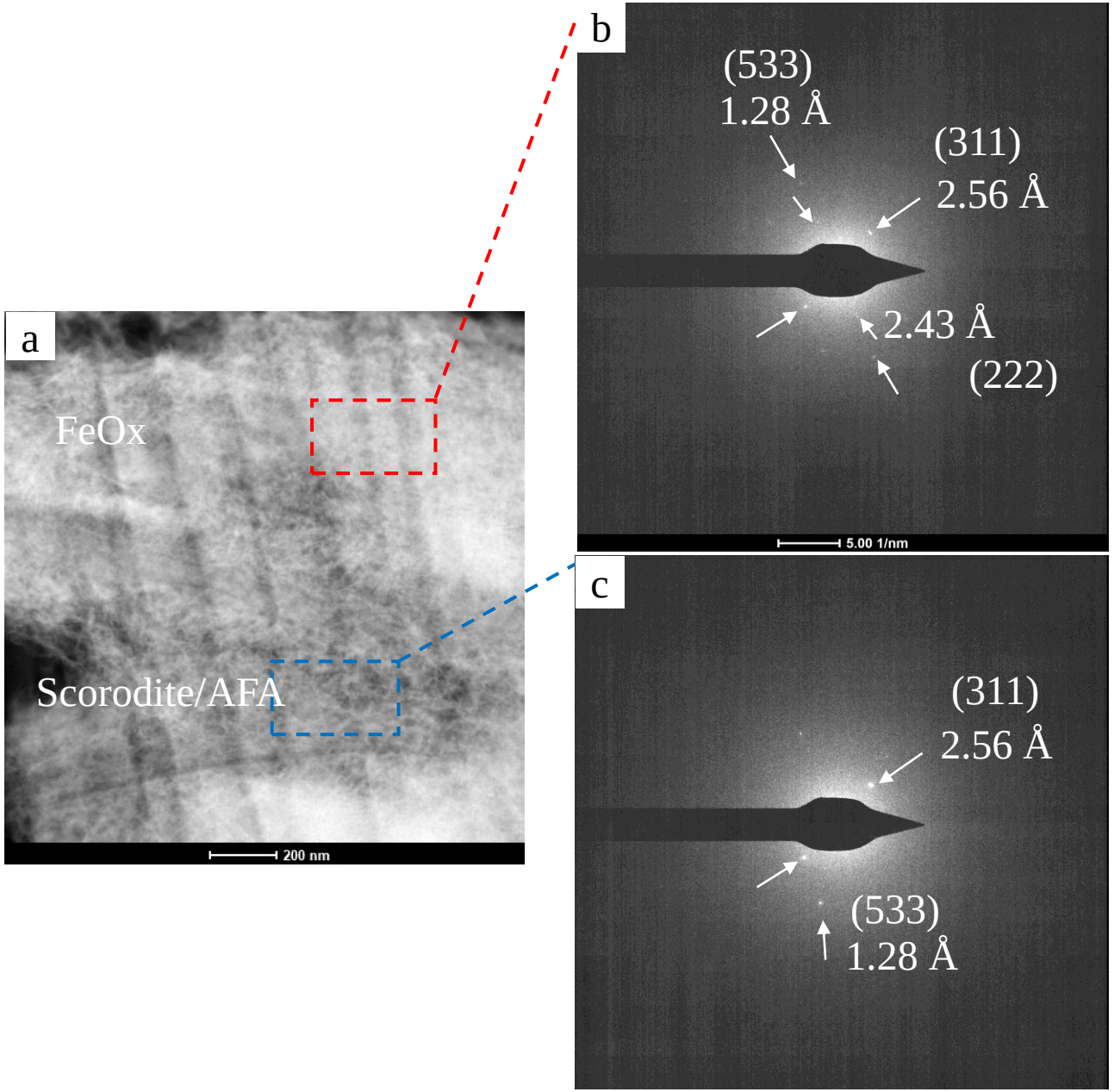

Figure S15: Unidentified Fe-(hydr)oxide (FeOx) - scorodite/AFA interface (a) HR-TEM image and (b)-(c) SAED pattern for scorodite/AFA

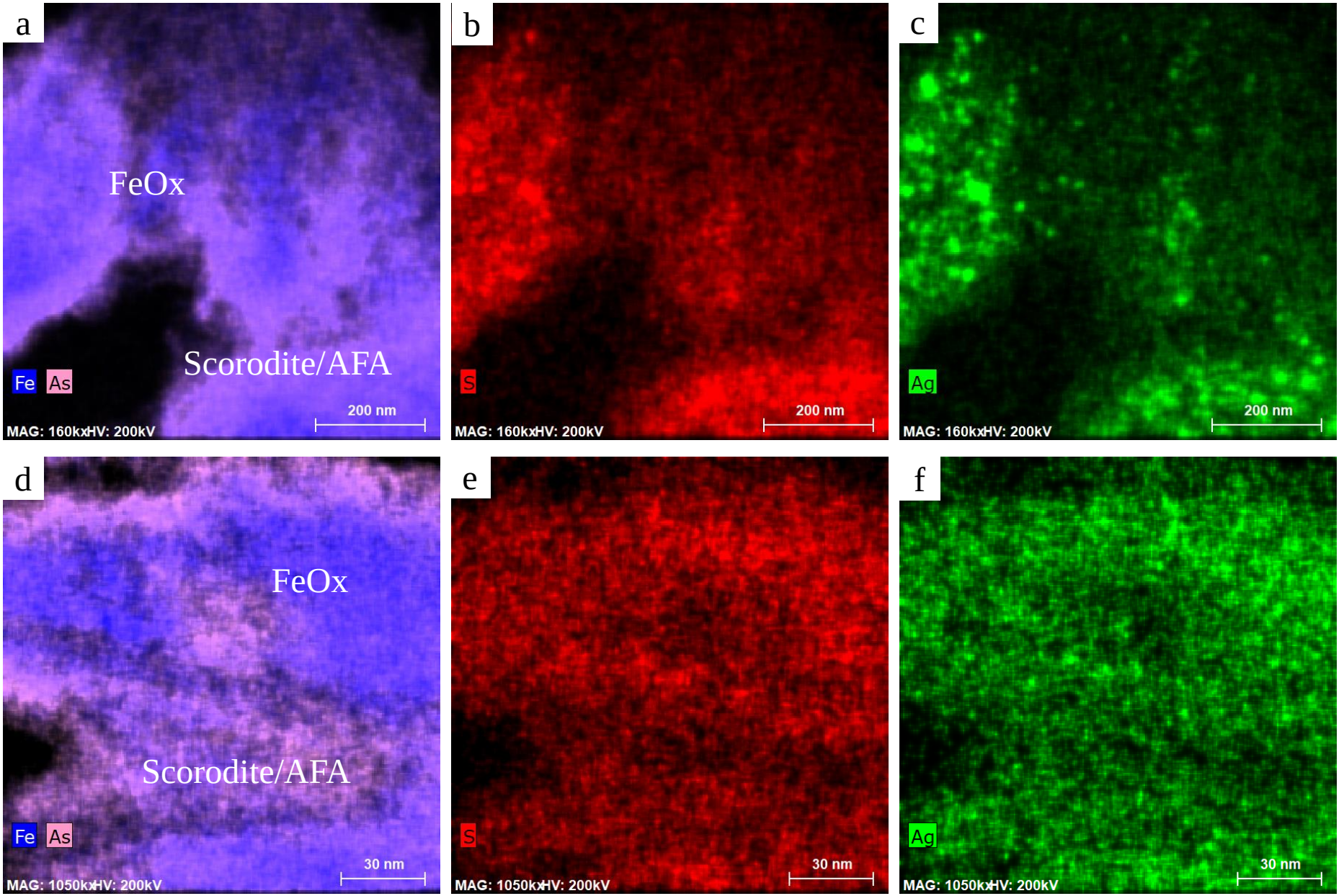

Figure S16: Unidentified Fe-(hydr)oxide (FeOx) and scorodite/AFA interface (a)-(f) STEM-EDS chemical distribution maps of Scorodite-bearing alteration layers on Fe-(hydr)oxides in TEM-FIB 2: (a)-(d) Fe and As; (b)-(e) S, (c)-(f) Ag;

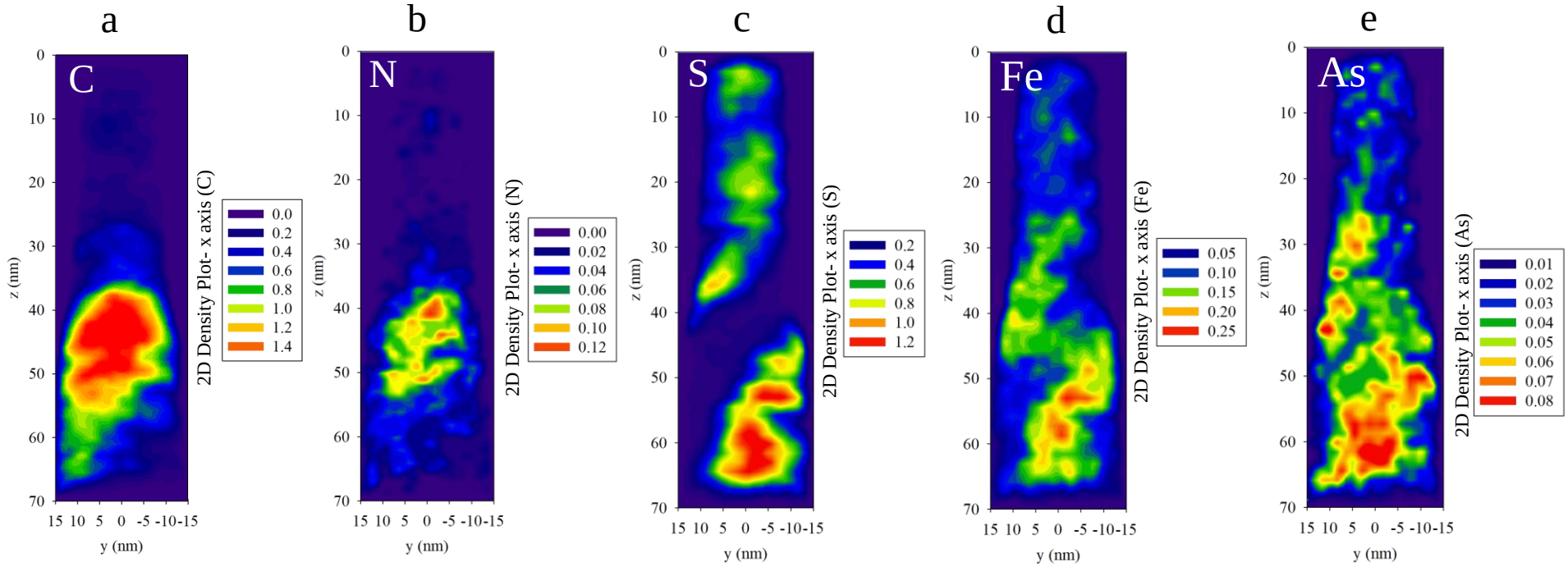

Figure S17: 2D heat maps for APT-FIB 1 (grain 1): (a) C, (b) N, (c) S, (d) Fe, (e) As