Cultivation reduces quantities of mineral-organic associations in the form of amorphous coprecipitates

Floriane Jamoteau[1,2,3,4*], Emmanuel Doelsch[2,3], Nithavong Cam[1], Clément Levard[1], Thierry Woignier[5,6], Adrien Boulineau[7], Francois Saint-Antonin[7], Sufal Swaraj[8], Ghislain Gassier[1], Adrien Duvivier[1], Daniel Borschneck[1], Marie-Laure Pons[1], Perrine Chaurand[1], Vladimir Vidal[1], Nicolas Brouilly[9], Isabelle Basile-Doelsch[1]

[1]Aix Marseille Université, CNRS, IRD, INRAE, Coll France, CEREGE, Aix-en-Provence, 13545, France
[2]CIRAD, UPR Recyclage et risque, Montpellier, F-34398, France
[3]Recyclage et Risque, Univ Montpellier, CIRAD, Montpellier, F-34398, France
[4]Institute of Earth Surface Dynamics, University of Lausanne, Lausanne, 1015, Switzerland
[5]Campus Agro Environnemental Caraïbes-IMBE-CNRS, B.P. 214, Petit Morne, Le Lamentin, Martinique, 97232, France
[6]Laboratoire Charles Coulomb UMR 5221 CNRS-UM2, Université Montpellier 2, Montpellier Cedex5, 34095, France
[7]Université Grenoble Alpes, CEA, LITEN, Grenoble, 38100, France
[8]Synchrotron SOLEIL, L'Orme des Merisiers, Departementale 128, Saint-Aubin, 91190, France
[9]Aix-Marseille Université, CNRS UMR 7288, IBDM, Marseille, 13000, France

Correspondence to: Floriane Jamoteau (floriane.jamoteau@unil.ch)

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Supplementary information 1. Mineral characterization of bulk soils and fine fractions of the forest and crop soils

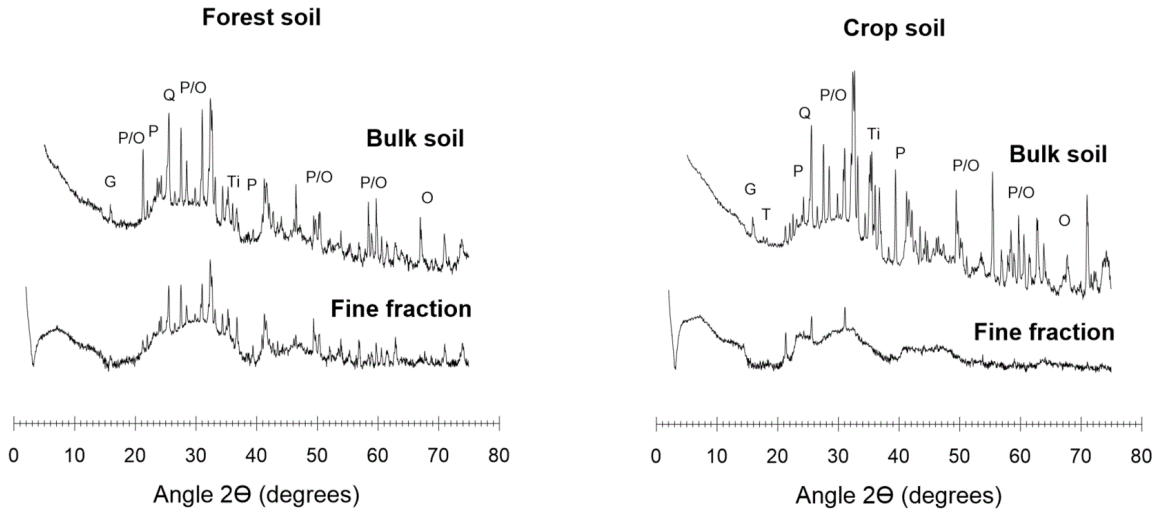

Figure S1. X-ray diffractograms comparing the bulk and fine fractions of forest and crop andosols. Displayed on a logarithmic scale, the mineral phases identified in the diffractograms are labeled as: plagioclase (P), orthopyroxene (O), gibbsite (G), tridymite (T), quartz (Q), and titanomagnetite (Ti). The broad diffusion bands suggest the presence of poorly crystallized or amorphous phases.

Supplementary information 2. Chemical mapping of mineral-organic associations with transects from crystalline to amorphous phases, using TEM-EDX.

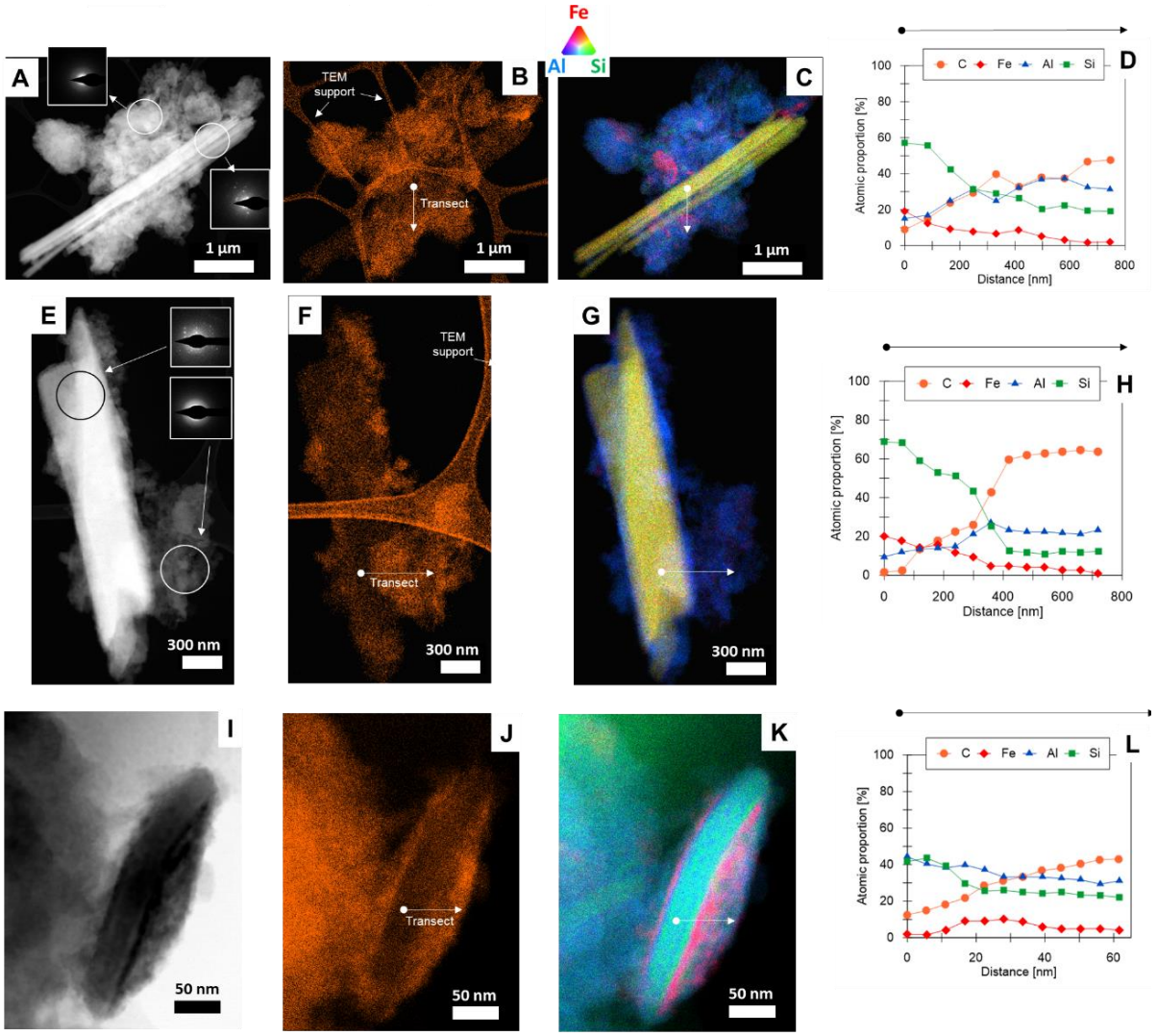

Figure S2. Chemical mapping of organo-mineral associations with transects from crystalline to amorphous phases, using TEM-EDX. The figures display: (A, E, I) TEM images highlighting fine soil minerals and the surrounding amorphous phase; (B, F, J) carbon distribution within these images; and (C, G, K) mappings of Al, Si and Fe. Transects show the atomic proportion transition from mineral to amorphous phase in (D, H, L). Mappings A-H were analyzed on the crop andosol, while mappings I-L were analyzed from the forest andosol.

Supplementary information 3. TEM images and high-resolution TEM images of the fine fractions of the forest and crop andosols

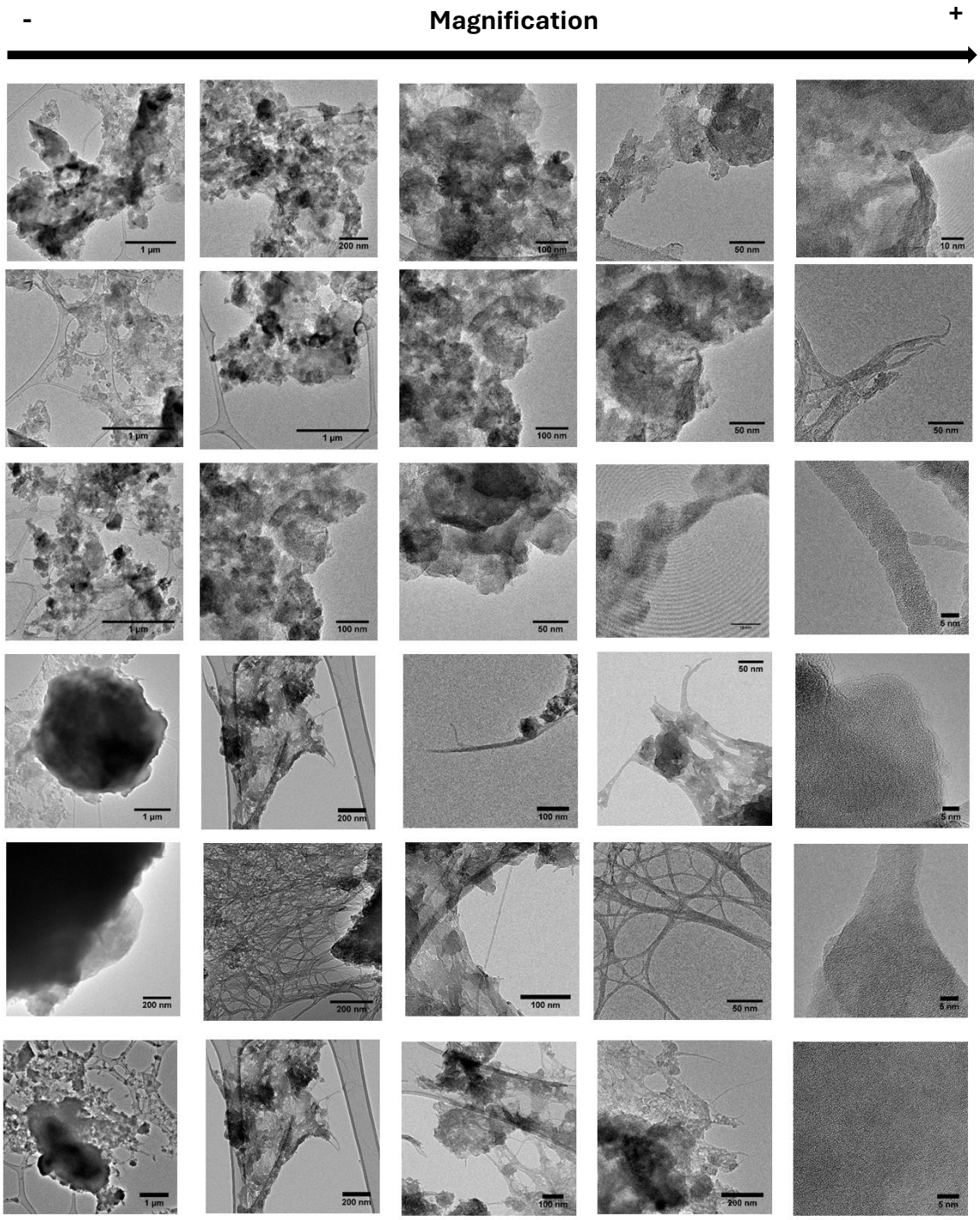

Figure S3.TEM and high-resolution TEM image analyses on the fine fractions of the forest andosol. Analyses were conducted in bright field from the micrometric to nanometric scale.

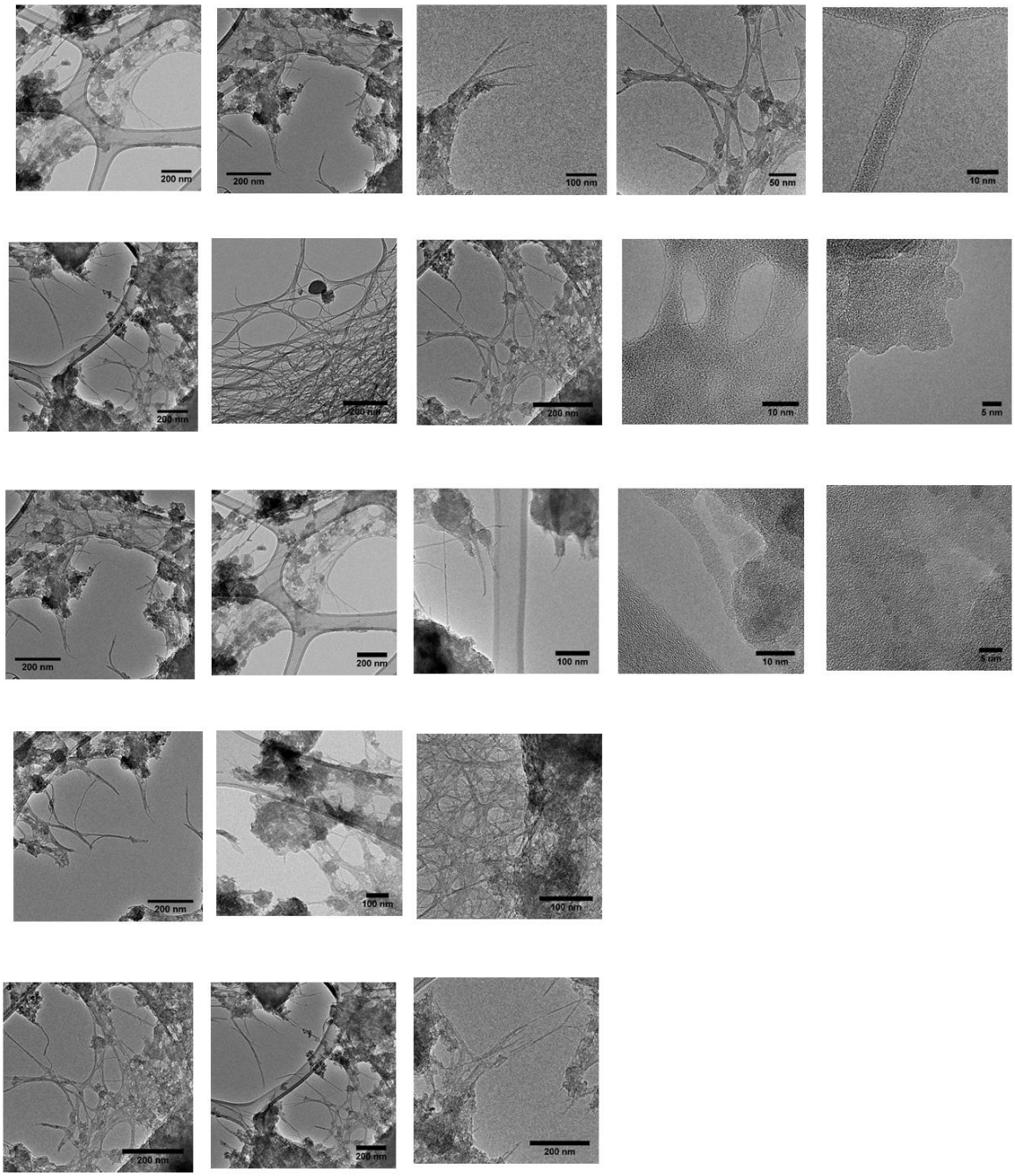

Figure S3. Continuation.

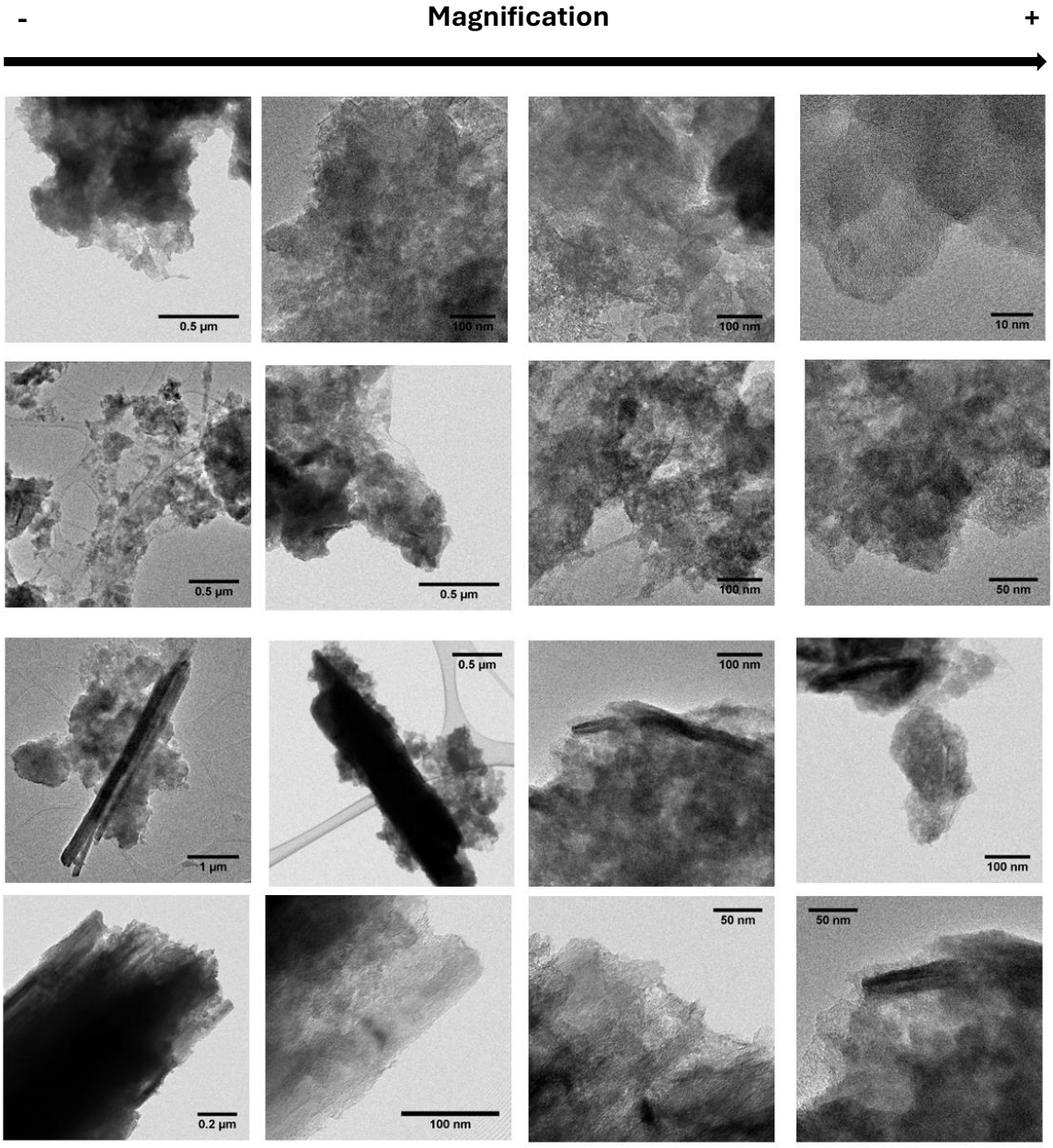

Figure S4.TEM and high-resolution TEM image analyses on the fine fractions of the crop andosol. Analyses were conducted in bright field from the micrometric to nanometric scale.

Supplementary information 4. TEM-EDX chemical mappings on the fine fractions of the forest and crop andosols

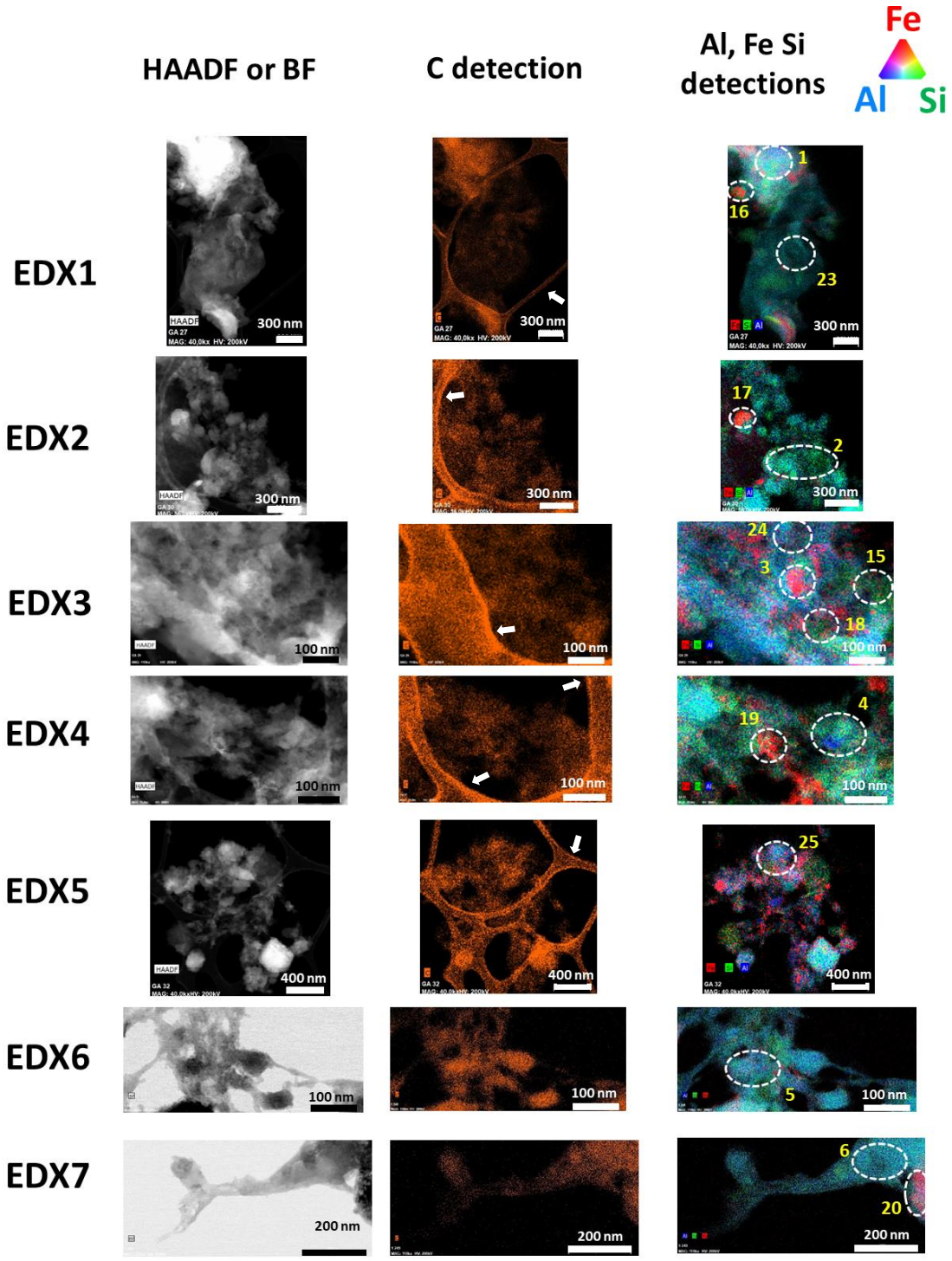

Figure S5. TEM-EDX chemical mapping of mineral-organic associations in fine fractions of the forest andosol. The images were acquired in dark-field (HAADF) and bright-field (BF) modes. In the C mappings, the arrows indicate the sample support (C lacey). In the Al, Fe and Si mappings, the numbered zones represent areas where atomic proportions were calculated (see proportions in Table S1).

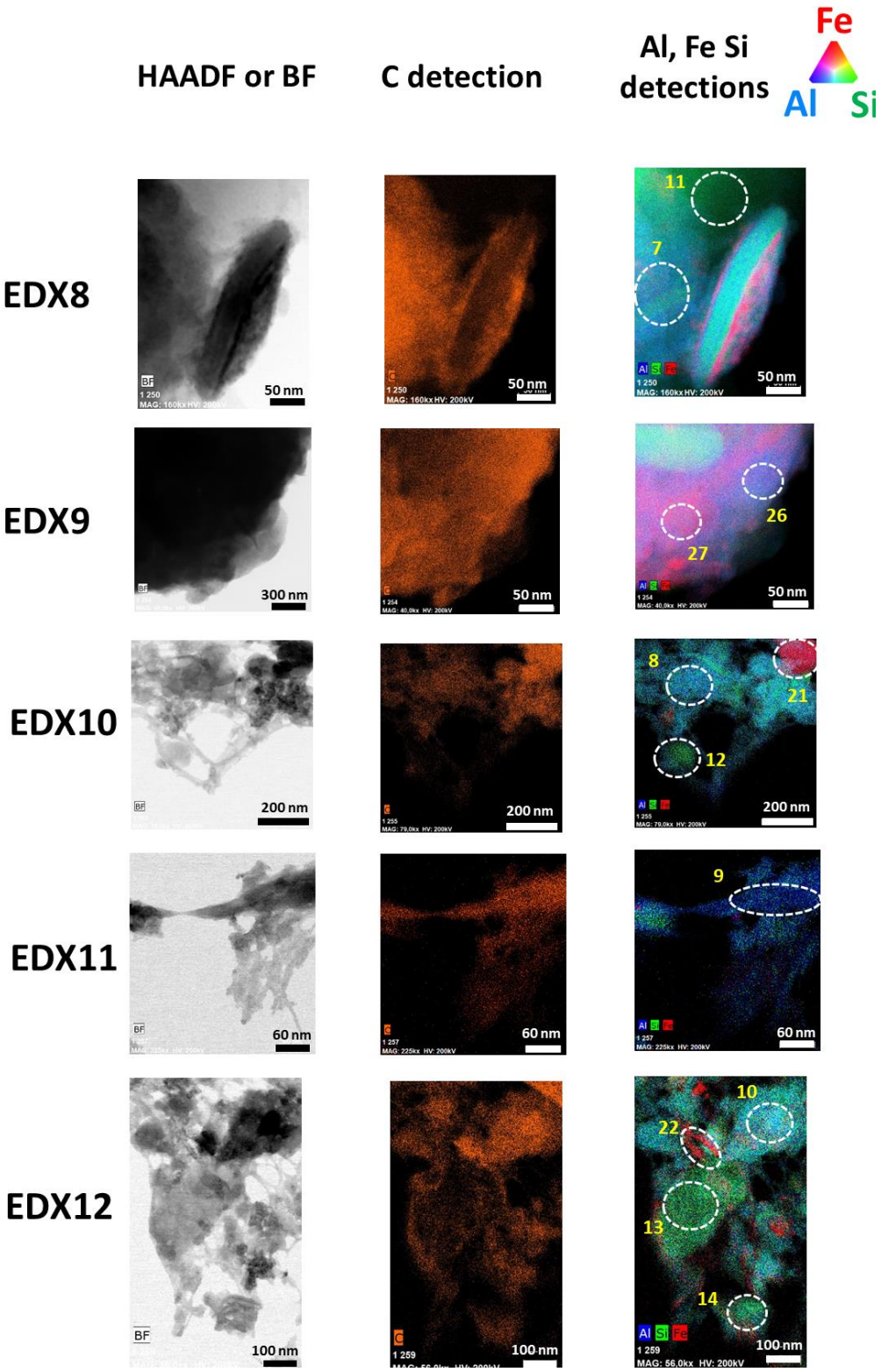

Figure S5. Continuation.

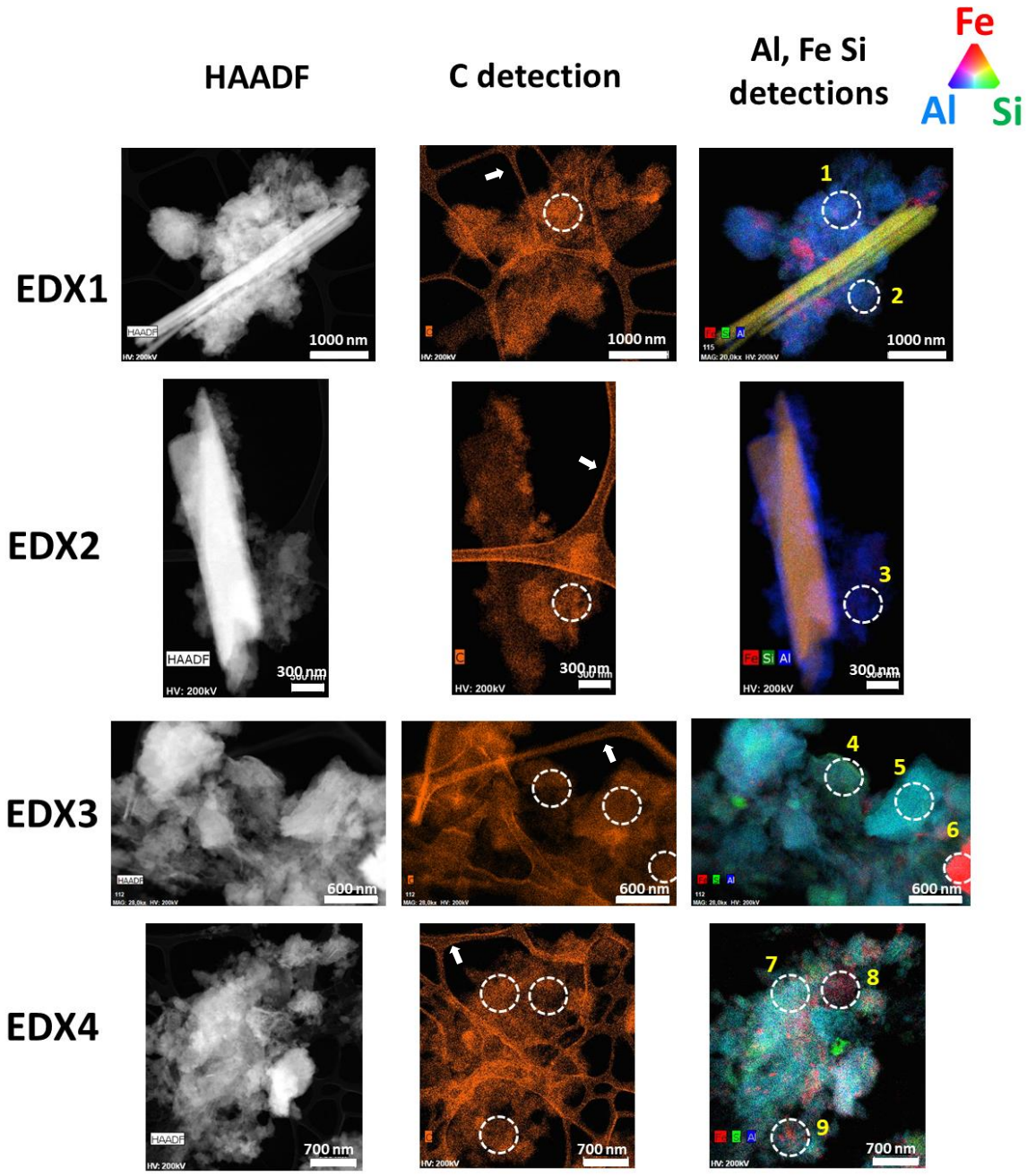

Figure S6. TEM-EDX chemical mapping of mineral-organic associations in fine fractions of the crop andosol. The images were acquired in dark-field (HAADF) mode. In the C mappings, the arrows indicate the sample support (C lacey). In the Al, Fe and Si mappings, the numbered zones represent areas where atomic proportions were calculated (see proportions in Table S2).

Table S1. Atomic proportions derived from TEM-EDX mappings of the forest soil. See the localization of individual areas in Fig. S5.

Area_number	EDX_number	Atomic proportions [%]						
		C	Fe	Al	Si	Ca	Mg	N
1	1	21.78	4.38	46.05	21.87	0.29	1.02	4.6
2	2	40.75	2.35	30.08	19.41	0.26	1.51	5.63
3	3	34.97	7.49	33.76	16.67	0.24	0.86	6
4	4	36.42	1.87	39.05	16.4	0.26	0.86	5.14
5	6	38.06	1.34	32.78	22.45	0.42	1.38	3.57
6	7	34.08	0.43	38.21	22.44	0.36	1.24	3.24
7	8	32.48	2.33	34.78	28.04	0.92	1.29	0.16
8	10	45.97	0.64	30.23	17.08	0.4	1.23	4.44
9	11	55.46	0.16	16.39	6.9	0.28	0.88	19.92
10	12	52.34	1.76	26.64	14.77	0.3	0.79	3.39
11	8	1.17	0.79	31.61	62.53	2.75	1.03	0.12
12	10	18.05	0.92	31.11	41.21	0.56	3.82	4.33
13	12	26.76	1.61	32.31	32.14	0.4	1.27	5.52
14	12	26.83	1.53	35.31	29.54	0.56	1.26	4.97
15	3	19.5	2.55	34.51	36.15	0.32	3.52	3.45
16	1	23.69	12.65	30.71	24.53	0.31	1.23	6.88
17	2	54.22	16.9	15.13	8.06	0.31	0.67	4.7
18	3	36.03	2.94	34.83	21.56	0.24	1.18	3.21
19	4	55.9	7.75	19.8	10.19	0.17	0.44	5.74
20	7	31.72	8.31	35.3	18.37	0.48	1.58	4.25
21	10	83.77	5.62	4.92	1.89	0.24	0.38	3.18
22	12	53.86	5.2	20.25	13.6	0.53	0.89	5.67
23	1	25.19	1.36	44.35	22.99	0.42	1.31	4.37
24	3	38.62	2.3	35.8	17.74	0.25	0.74	4.55
25	5	44.73	1.56	31.75	15.23	0.32	0.77	5.64
26	9	13.16	5.76	52.44	27.14	0.33	0.79	0.38
27	9	13.84	4.4	51.93	27.09	0.3	0.95	1.49

Table S2. Atomic proportions derived from TEM-EDX mappings of the crop soil. See the localization of individual areas in Fig. S6.

Area_number	EDX_number	Atomic proportions [%]						
		C	Fe	Al	Si	Ca	Mg	N
1	1	29,5	3,2	40,3	22,2	0,5	1,2	3,2
2	1	39,1	2,6	34,2	18,7	0,5	0,9	4,2
3	2	52,6	3,0	26,6	13,9	0,3	0,7	3,1
4	3	21,2	3,2	40,4	27,6	0,3	2,5	4,8
5	3	13,6	0,7	52,6	30,6	0,3	0,8	1,5
6	3	13,7	49,9	22,3	12,3	0,2	0,3	1,2
7	4	24,6	2,2	44,2	25,2	0,4	1,3	2,0
8	4	48,7	3,2	29,5	13,7	0,3	0,9	3,7
9	4	43,5	2,7	29,8	15,5	0,3	0,9	7,4

Supplementary information 5. TEM-EELS chemical mappings on the fine fractions of the forest and crop andosols

To investigate the colocalization of C with elements derived from mineral weathering (±Fe, ±Al, and ±Si) down to the nanometer scale, we performed TEM-EELS (Transmission Electron Microscopy - Electron Energy Loss Spectroscopy) chemical mapping on the andosol fine fractions. Our results showed that in the electron amorphous phase, C was predominantly found in elemental mixtures (Fig. S7, S8). In the forest soil mapping, 54% of C colocalized with a C+Al+Si elemental mixture, while 37% colocalized with Al alone. Considering that this technique tends to underestimate Si detection compared to Al and Fe detections (Jamoteau et al., 2023), we estimate that the majority of C was colocalized in a C+Al+Si mixture. In the crop soil mapping, results showed that 93% of C was associated with elemental mixtures made of C+Al+Fe+Si, C+Al+Fe, or C+Al+Si (Fig. S8), reaffirming that C is present in elemental mixtures within andosols at scales below 15 nm.

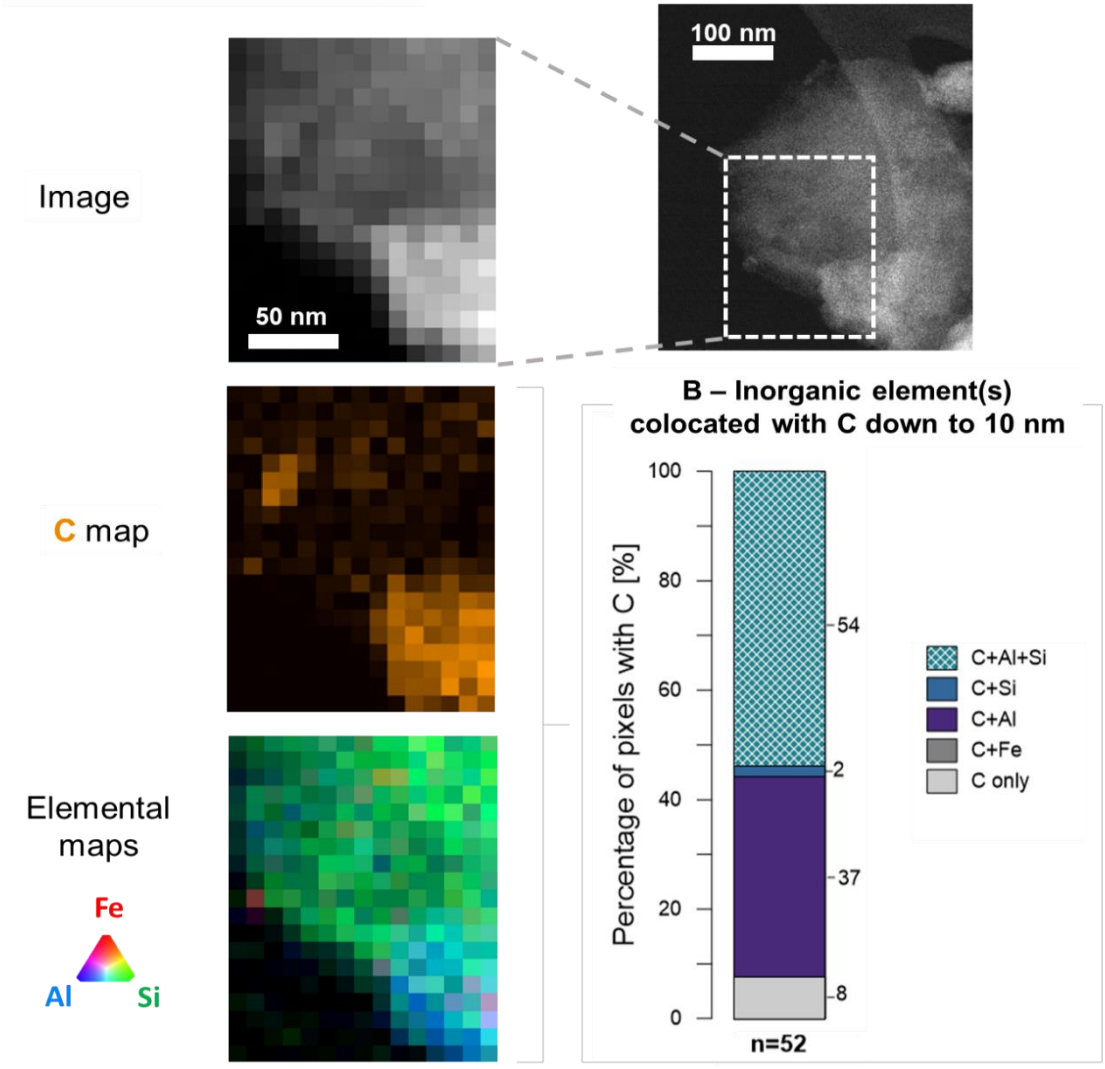

Figure S7. Chemical map on fine fractions of the forest soil using TEM-EELS mapping (pixel size of 10 nm). (A) images, C mapping and Fe, Al and Si mapping. (B) Compilation of collocated elements (Fe, Al and Si) with C in 10-nm-pixels-size. *n* stand for the number of pixels with a C detection (n=52).

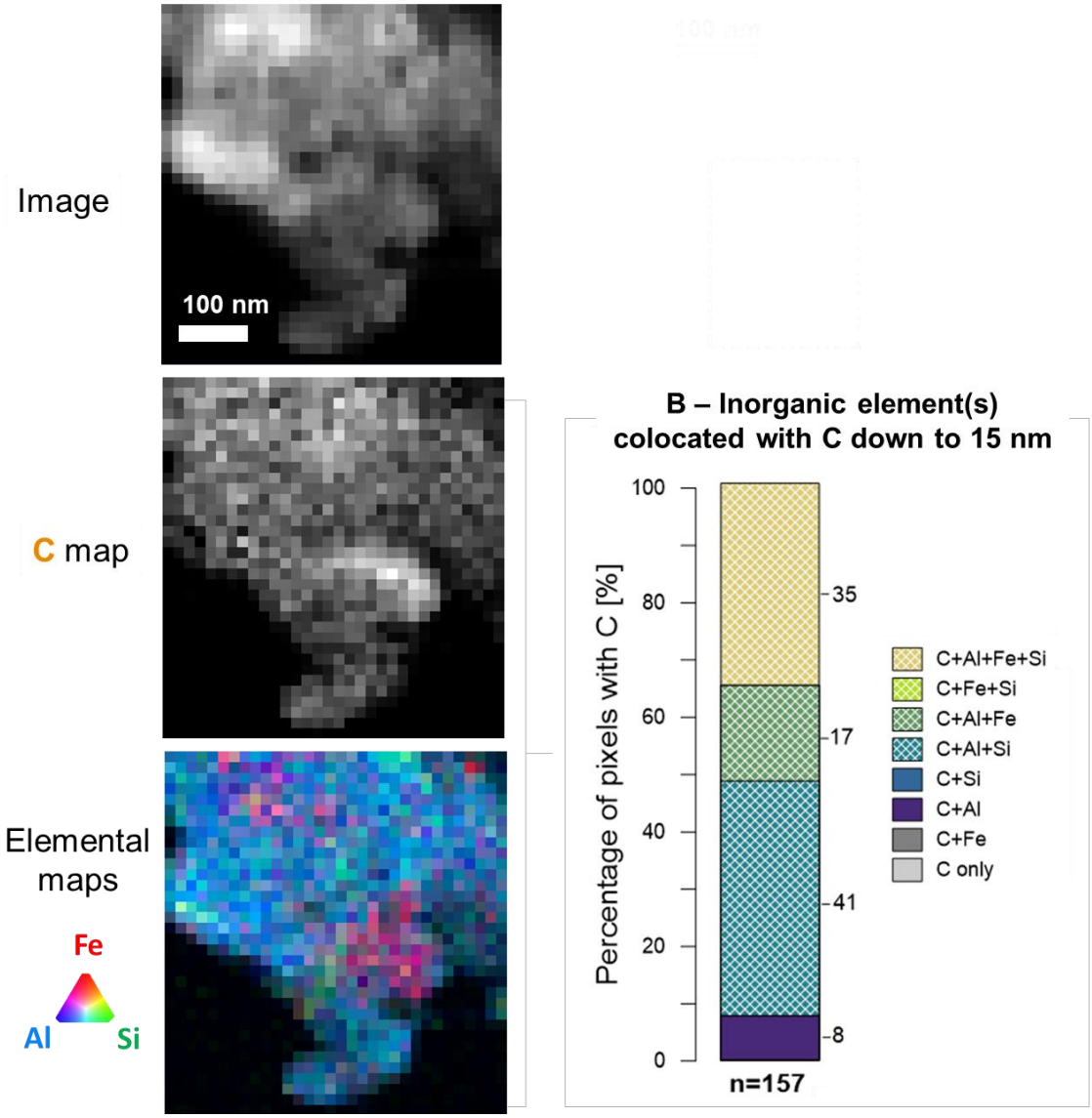

Figure S8. Chemical map on fine fractions of the crop soil using TEM-EELS mapping (pixel size of 15 nm). (A) images, C mapping and Fe, Al and Si mapping. (B) Compilation of collocated elements (Fe, Al and Si) with C in 10-nm-pixels-size. *n* stand for the number of pixels with a C detection (n=157).

Supplementary information 6. STXM mappings on the fine fractions of the forest and crop andosols

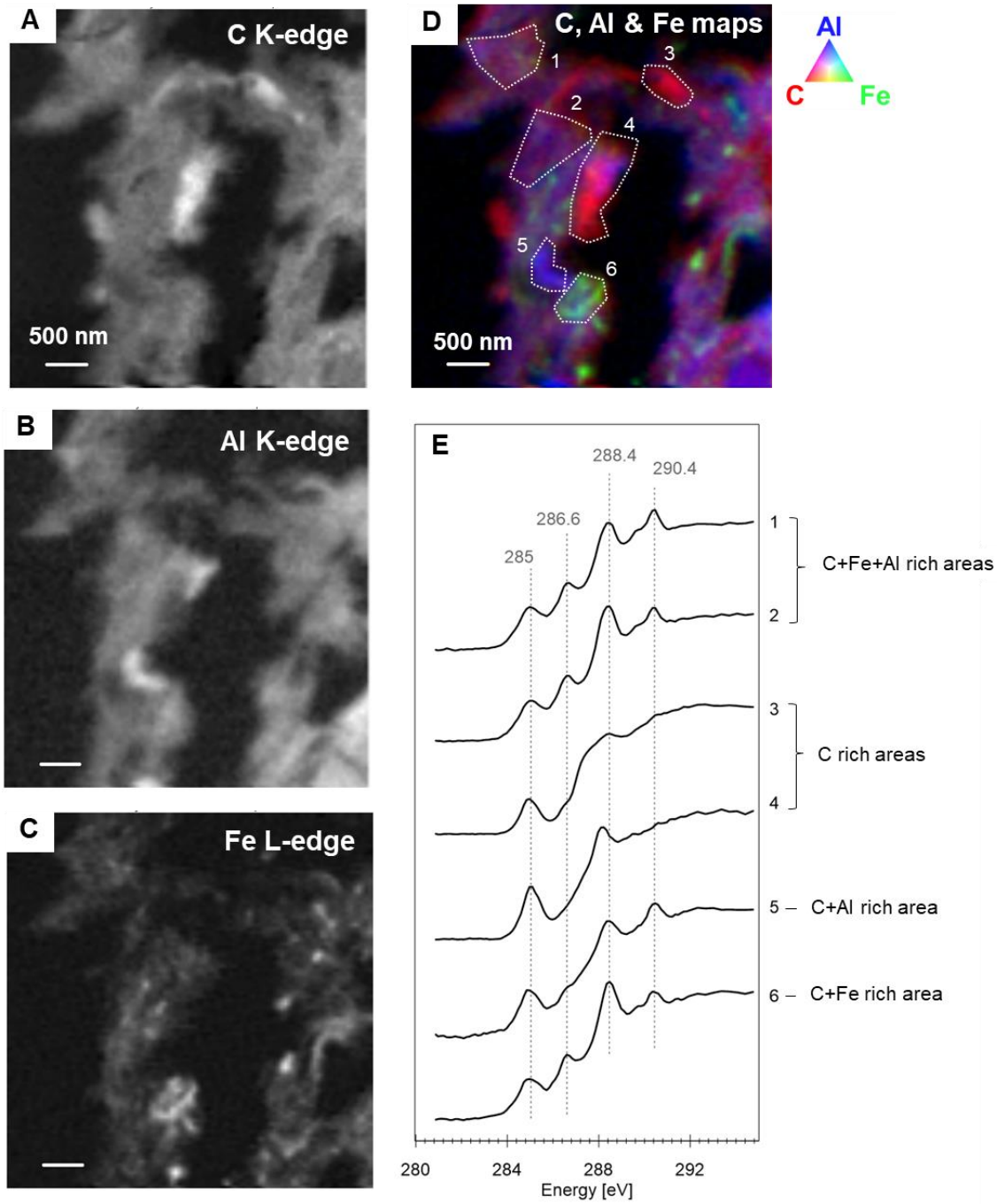

Figure S9. Chemical mapping and organic matter characterization within the forest soil fine fractions, using STXM mapping. (A) STXM chemical mappings at the C K-edge, (B) Al K-edge, and (C) Fe L-edge. (D) RGB composite image of the C K-edge, Al K-edge and Fe L-edge. (E) C K-edge spectra of the delineated area (outlined in panel D) showcasing principal energy bands associated with aromatic C=C and C=H (~285 eV), phenolic C-OH and ketonic C=O (~286.6 eV), carboxylic C=O and C-OH (~288.4 eV), and carbonyl C=O (~290.4 eV).

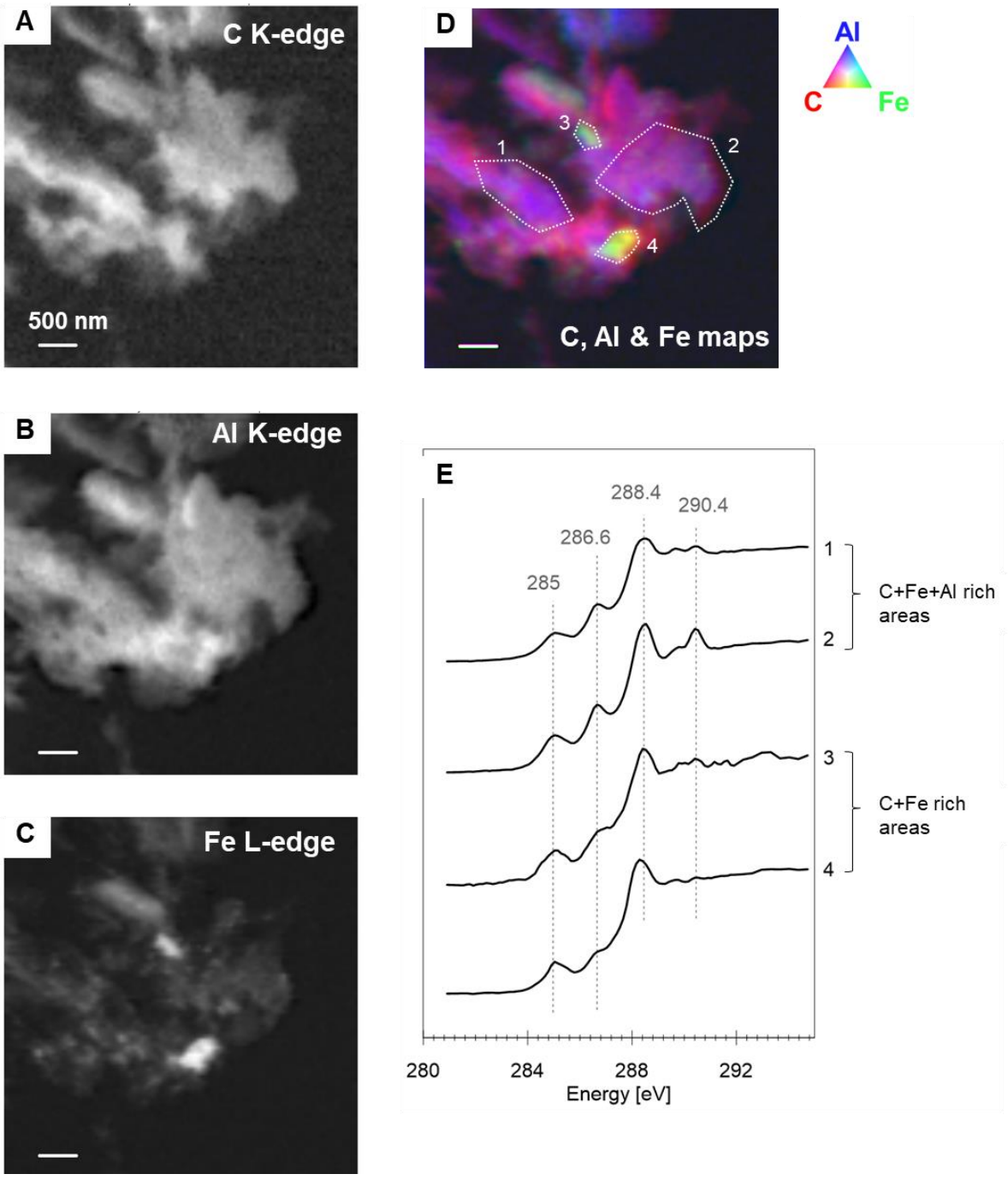

Figure S10. Chemical mapping and organic matter characterization within the crop soil fine fractions, using STXM mapping. (A) STXM chemical mappings at the C K-edge, (B) Al K-edge, and (C) Fe L-edge. (D) RGB composite image of the C K-edge, Al K-edge and Fe L-edge. (E) C K-edge spectra of the delineated area (outlined in panel D) showcasing principal energy bands associated with aromatic C=C and C=H (~285 eV), phenolic C-OH and ketonic C=O (~286.6 eV), carboxylic C=O and C-OH (~288.4 eV), and carbonyl C=O (~290.4 eV).

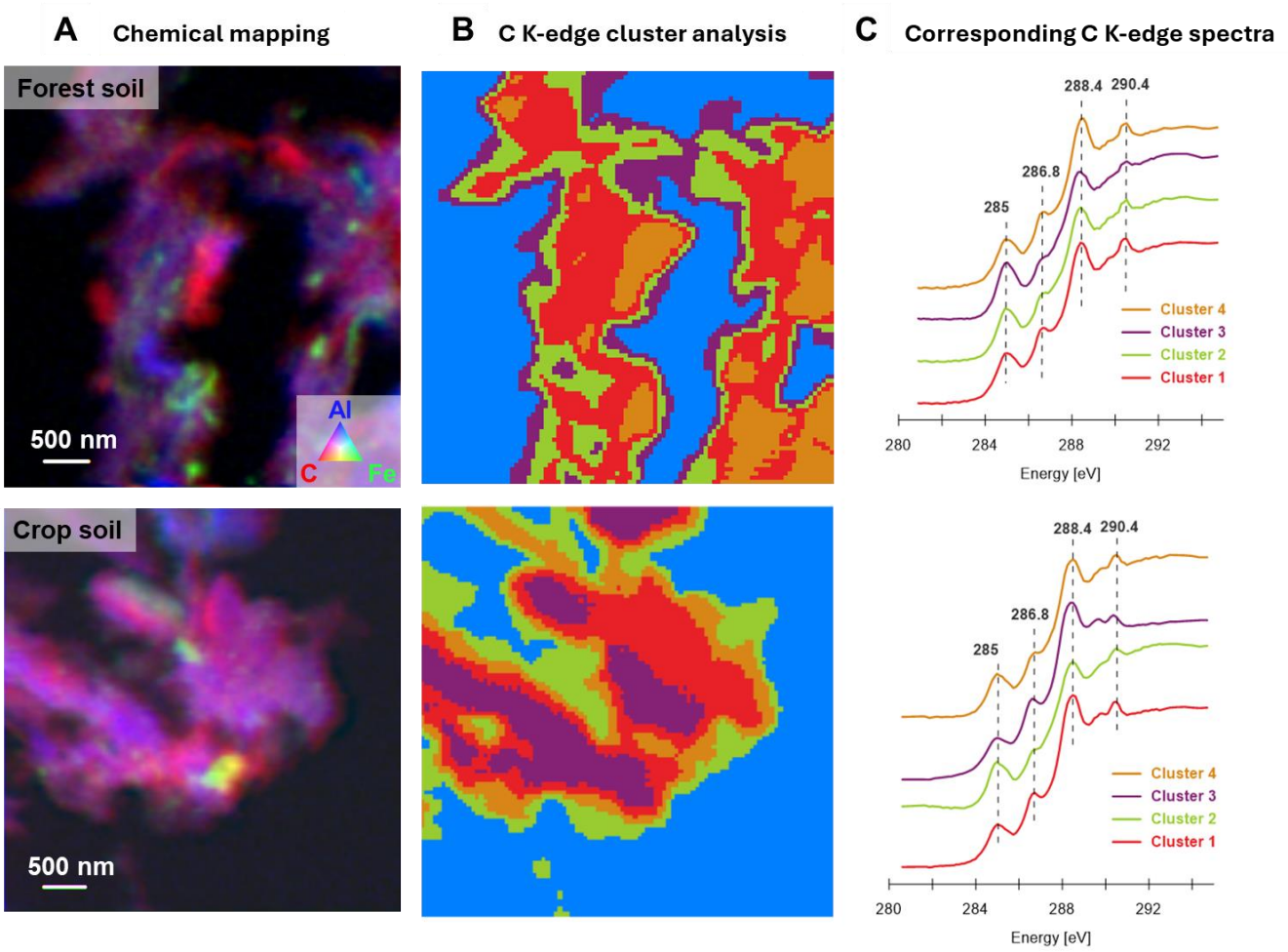

Figure S 11. Cluster analysis of the C K-edge of in forest and crop soil fine fractions. (A) RGB composite images showing the chemical mapping of the C K-edge, Al K-edge, and Fe L-edge. (B) Cluster analysis of the C K-edge. (C) Corresponding average C K-edge spectra for the identified clusters.