Supplementary information (SI)

Thickness calculation from sputtering.

For 100 s of sputtering,

$$Q = I * t$$

I is the current, which is 10 nA in this study, and the time (t) is 100 s.

Thus,

$$Q = 100*10*10^{-9}A*6.25*10^{18}/(2.5\ mm*2.5\ mm) = 1*10^{14}\ ions\ cm^{-2}$$

To determine the sputtering thickness, the sputtering efficiency is needed. We compared published data reporting similar measurement conditions(1), and the sputtering efficiency (slope) is correlated with the voltage (Figure S9). We used the fitting equation to determine that the efficiency at 4 eV is 0.22 μm $*10^{-15}$ $ions^{-1}$ cm^2. Therefore, the thickness can be assessed as followed:

$$Thickness_{100s} = 0.22\ \mu m*10^{-15}\ ions^{-1}cm^2*1*10^{14}\ ions\ cm^{-2} = 20\ nm$$

Considering the distinct difference between the organic leucine film and the soil mineral-OM associates, we roughly assessed the thickness removed by 100 s, 600 s and 1300 s of sputtering as approximately 20 nm, 130 nm and 280 nm, respectively.

Molecular component modelling of the OM

Composition modelling of the OM at the SWI was based on C 1s speciation carried out by XPS and principally described by Gerin (1993)(2). Briefly, the components of the OM were simplified as protein, glucans and lipid substances. Because the N 1s component was mainly amides, the N/C ratio was a measure of the protein content. The C=O/C and C-(O, N)/C ratios resulted from proteins and glucans, and the C=O/C and C-(O, N)/C ratios resulted from glucans. Minorly modified equations established by using the atom concentration ratios relative to carbon are as follows: $(N/C)_{observed} = 0.27\ C_{protein}$ (1); $[C\text{-}(O, N)/C]_{observed} = 0.32\ C_{protein} + 0.833\ C_{glucans}$ (2); $C_{lipid} = 1 - C_{protein} - C_{glucans}$ (3). $C_{protein}$, $C_{glucans}$, and C_{Lipd} are the proportions of the total surface carbon associated with each molecular constituent.

Figures

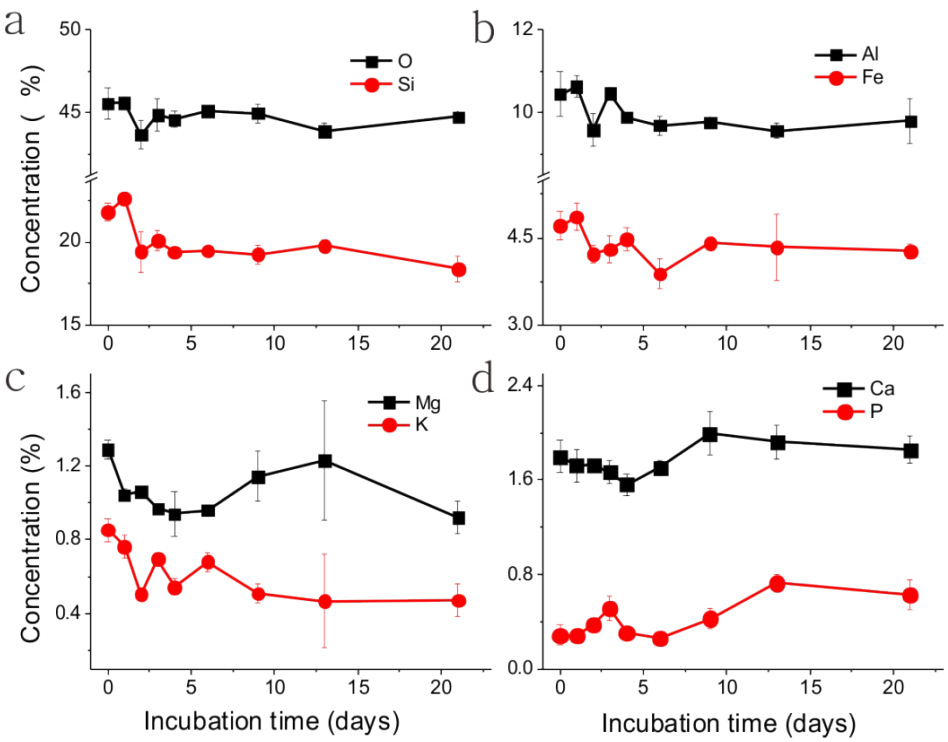

Figure S1: Dynamics of main elements O, Si, Al, Fe, Mg, Ca, K and P at the Mollisol surface during the 21 days of incubation.

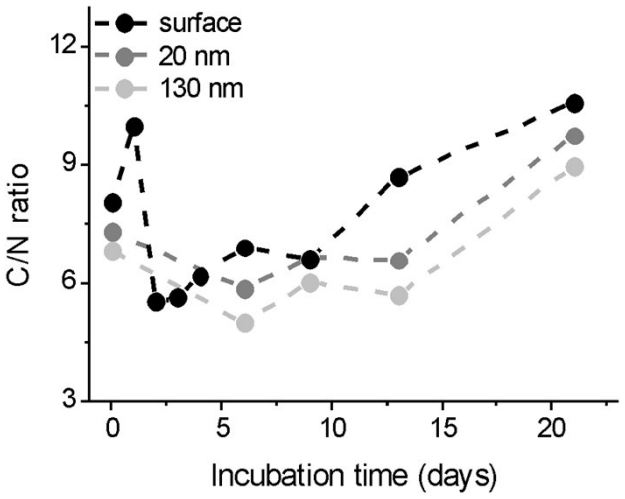

Figure S2: Depth profile of C/N ratio in OM during the formation of the new SWIs.

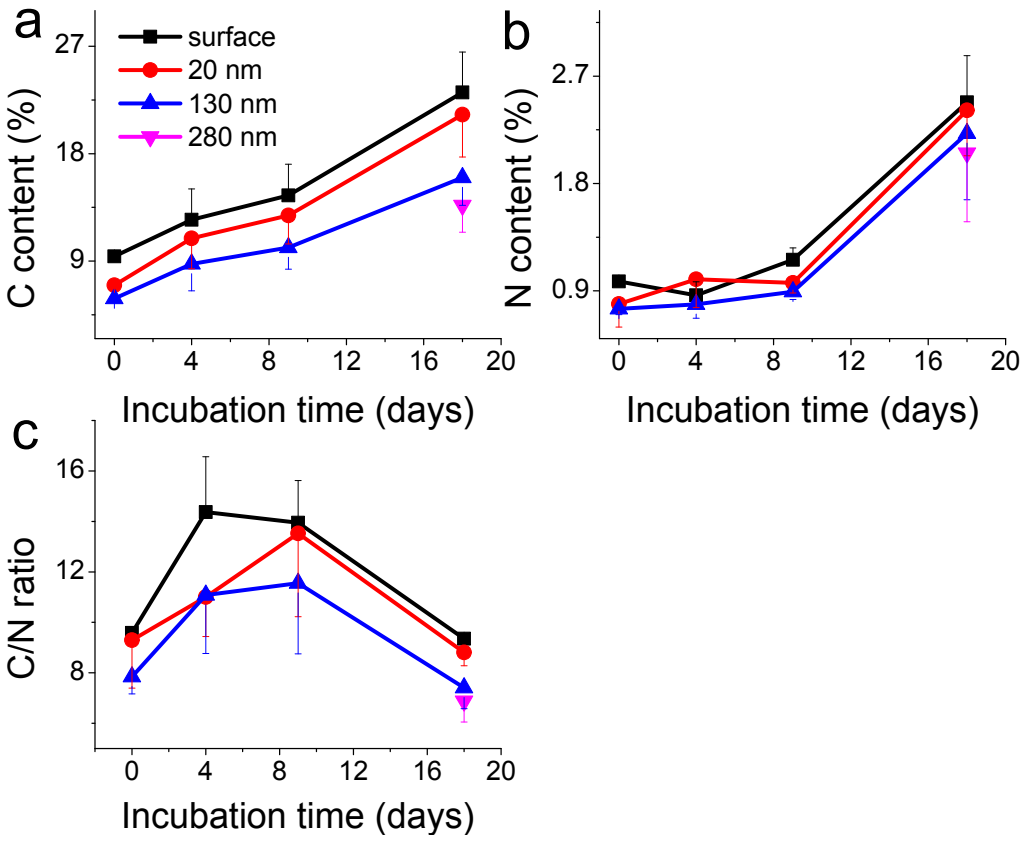

Figure S3: Dynamic profiling of C (a), N (b) and the C/N ratio (c) at the sterile soil interface by 100 s, 600 s and 1100 s of sputtering during the 18 days of incubation.

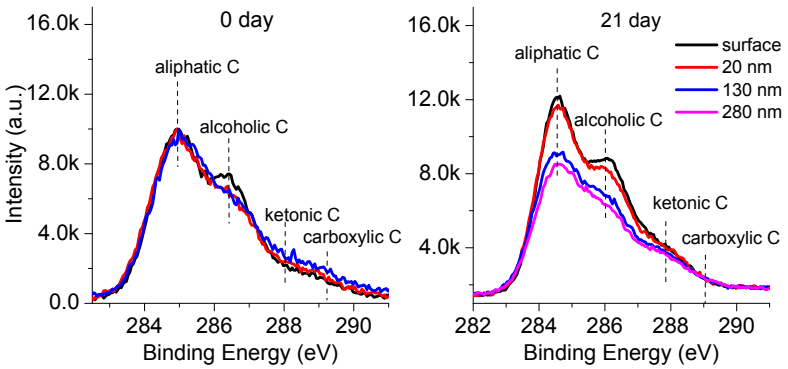

Figure S4: Depth profiles of the C 1s spectrum at the sterile soil interfaces at the start and after 21 days of incubation. The interfaces were exposed by continuous sputtering (surface, 20 nm, 130 nm, 280 nm) and XPS measurement. Four typical carbon species (aliphatic C, alcoholic C, ketonic C and carboxylic C) were identified.

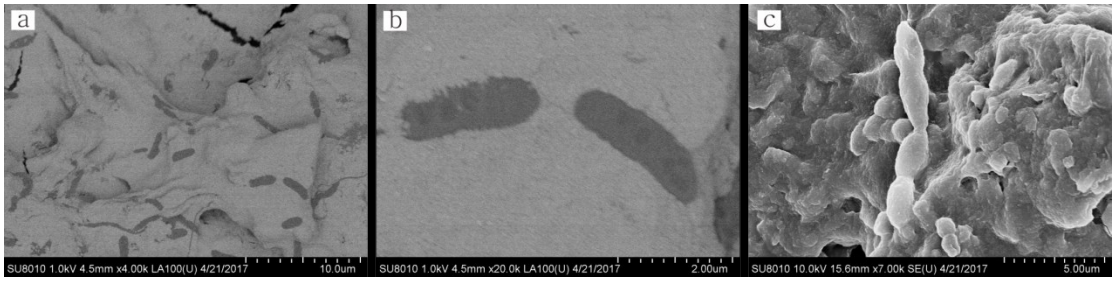

Figure S5: Microorganisms at the Mollisol BGI. (a) and (b) Backscatter electron imaging of the soil dots on the SoilChip at 10 μm and 2 μm scales, respectively. The black stick-like objects are carbon-rich microorganisms. (c) Scanning electron microscopy (SEM) image of the stick-like objects on the soil surface, confirming that these objects are microorganisms.

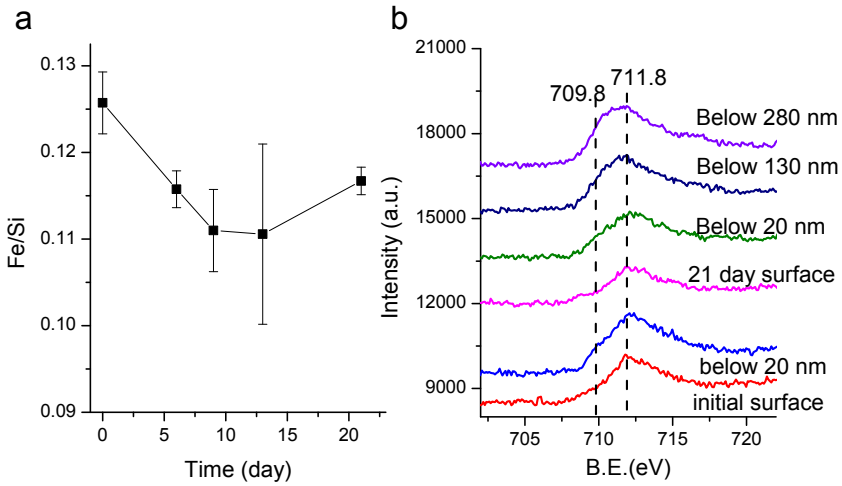

Figure S6 Changes in the Fe/Si ratio and the Fe 2p spectrums during the formation of the soil-water interfaces.

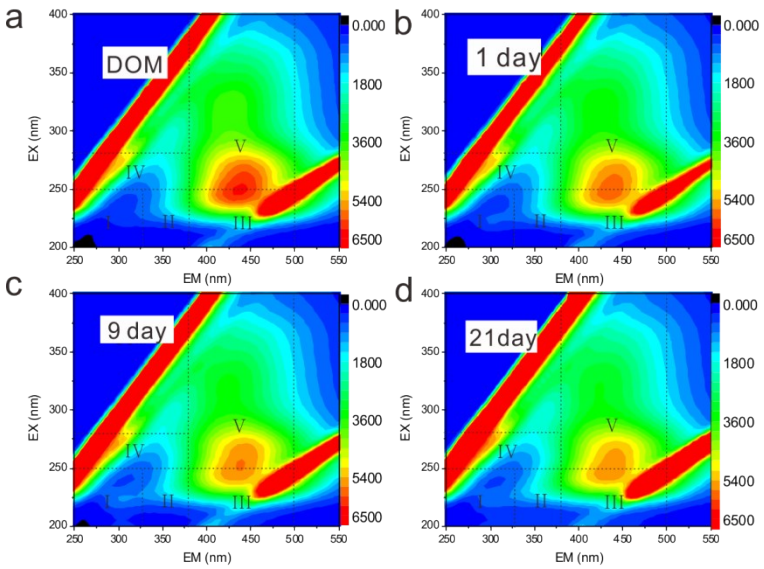

Figure S7: 3D-EEM spectra of a solution on the SoilChip. EEM maps were divided into five regions(3, 4): region I (Ex 200–250 nm, Em 250–330 nm) was related to tyrosine-like protein, region II (Ex 200–250 nm, Em 330–380 nm) was related to tryptophan-like protein, region III (Ex 200–250 nm, Em 380–500 nm) was related to phenol or polysaccharides-like organics, region IV (Ex 250–280 nm, Em 200–380 nm) was related to soluble microbial by-product-like materials, and region V (Ex 250–400 nm, Em 380–500 nm) was related to modified lignin-like organic matter. (a) The spectra of original DOM extracted from the Mollisol. (b), (c) and (d) are the spectral maps of the solution after 1, 9 and 21 days of incubation.

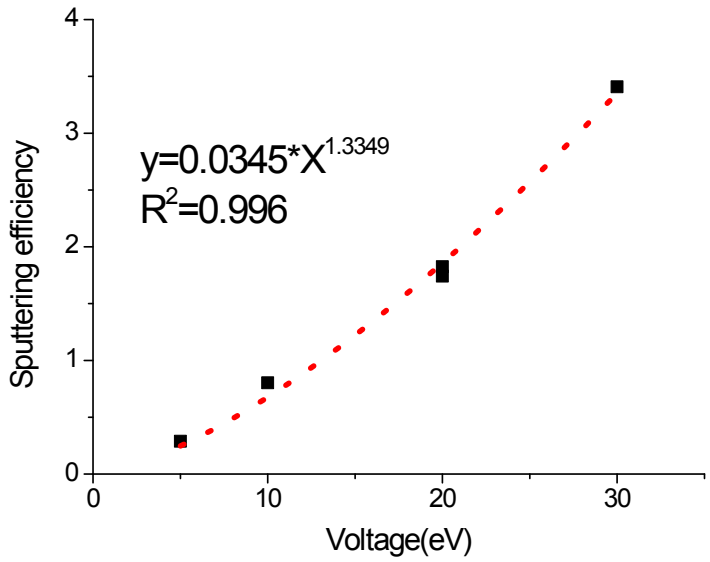

Figure S8: Correlation between sputtering efficiency (μm (10^{15} ions $cm^2)^{-1}$) and voltage. Data is from the K. Ichiki et al. (2008). The sputtering efficiency is calculated as the slope between the sputtering depth of leucine films and sputtering fluence with 5–30 keV Ar cluster ions.

Tables

Table S1: Percentage of four fitted carbon species at the BGI surface after 21 days and 600 s of sputtering.

Sputtering time	0s	600s
aliphatic C	33.8%	41%
alcoholic C	47.9%	37.7%
ketonic C	14.7%	14%
carboxylic C	3.5%	7.2%

Table S2: Composition of the OM at different depths after 21 days of incubation.

depth (nm)	C (%)	N (%)	P (%)	C/N	C/P
surface	16.5	1.3	0.7	12.5	22.7
20	11.1	0.9	0.3	12.7	37.1
60	9.6	1.0	0.6	10.1	15.0
130	8.9	0.9	0.5	9.9	16.5
240	7.9	1.2	0.9	6.6	10.2
280	8.0	0.9	0.7	8.8	11.0

References

1. Ichiki K, Ninomiya S, Nakata Y, Honda Y, Seki T, Aoki T, et al. High sputtering yields of organic compounds by large gas cluster ions. Applied Surface Science. 2008;255(4):1148-50.
2. Gerin PA, Dufrêne Y, Bellon-Fontaine M, Asther M, Rouxhet P. Surface properties of the conidiospores of Phanerochaete chrysosporium and their relevance to pellet formation. Journal of bacteriology. 1993;175(16):5135-44.
3. Sun J, Guo L, Li Q, Zhao Y, Gao M, She Z, et al. Three-dimensional fluorescence excitation–emission matrix (EEM) spectroscopy with regional integration analysis for assessing waste sludge hydrolysis at different pretreated temperatures. Environmental Science and Pollution Research. 2016;23(23):24061-7.
4. Zhu L, Qi H-y, Kong Y, Yu Y-w, Xu X-y. Component analysis of extracellular polymeric substances (EPS) during aerobic sludge granulation using FTIR and 3D-EEM technologies. Bioresource technology. 2012;124:455-9.