Characterization and formation of the pristine rhizoliths around

Artemisia roots in dune soils of Tengeri Desert, NW China

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ABSTRACT

Rhizoliths are the products of mineralization, petrification, or fossilization around and/or within plant roots. Among them, carbonate rhizoliths are the most common. Pristine carbonate rhizoliths with co-existing plant root relicts in the Tengeri Desert, NW China were studied, with a combination of intensive field observations and laboratory methods such as microscopy, scanning electronic microscopy, energy dispersive X-ray spectra, radiocarbon dating, and isotope mass spectrometer. The field observations revealed that the pristine rhizoliths are only present at the sites where *Artemisia sphaerocephala* Krasch are growing i.e. in swales among sand dunes. Soil moisture of the swales is the main controlling factor of rhizoliths

formation. It is in turn affected by the soil physical properties, landscape position, and climate variability, consistent with the locations of sampled rhizoliths in the swales where *Artemisia* plants are mostly distributed. The ^{14}C AMS dating indicated that the rhizoliths are much older (4000-5000 years) than their co-existing modern plant root relicts in agreement with field observations. Morphological, mineralogical and isotopic analyses revealed that carbon sources used for the rhizoliths formation were partially derived from decomposing plant roots but with significant contribution from dissolution of lithogenic carbonates. The calcium sources were suggested to be the in situ weathering of minerals (mostly lithogenic carbonates) and the pressure-dissolution of carbonates. Enough CO_2 from the root decomposition have triggered carbonate accumulation around the root to form rhizoliths. Other minor chemical components of the root are S, N, P, which produce acidic water with the negative ions of SO_4^{2-} , NO_3^- , PO_4^{3-} , have also favored acidic soil environment and enhanced carbonate dissolution and mineral weathering. Redox environment around *Artemisia* roots were also observed to be a key factor for the pristine rhizolith formation. The pristine rhizoliths were preferentially formed in semi-closed redox condition with water nearly always available at intermediate depths. In addition, they were formed through carbonate epidiagenesis in shallow soils of the desert. Altogether, our results showed that the formation of the pristine rhizoliths was affected by the combination of several environmental factors. This led us to propose a conceptual model of rhizolith formation in desert soils.

Keywords:

Pristine rhizoliths; Dune soil; Redox condition; *Artemisia* roots; Soil moisture; Carbonatization

1. Introduction

Rhizoliths are defined as organo-sedimentary structures produced through accumulation and/or cementation of mineral matters such as carbonates around or within vascular plant roots (Klappa, 1980). They are products of interaction between roots and soils/sediments, and occur mostly as trace fossils without showing root morphological structure (Genise, 2017; Buatois & Mángano, 2011). But, occasionally rhizoliths occur in old strata as body fossils for petrified roots (Klappa, 1980). In paleontology and taphonomy, rhizoliths are also called fossil roots formed through fossilization, which comprises the processes leading to the preservation of trace of life in the geological record (Li et al., 2013). Rhizoliths are important materials that can be used to study paleoclimate (Li et al., 2015a, b), paleoecology (Nascimento et al., 2019; Khechai & Daoud, 2016) and paleohydrology (Bojanowski et al., 2016; Kraus & Hasiotis, 2006), as well as paleosols identification (Barta, 2011; Retallack, 2001) and carbon cycle (Alonso-Zarza, 2018).

Although many types of carbonate rhizoliths have been found since the Silurian geological period, their formation mechanisms have been widely debated with many hypotheses brought forward (Sun et al., 2019a,b; Li et al., 2018; Alonso-Zarza et al., 2018). Several related questions remain open, such as the relationship between

vegetation types and rhizolith formation, the chemical and morphological differences between carbonate encrustations around living and/or deceased roots and the relationship between modern rhizosheaths and rhizoliths.

So far, no pristine rhizoliths around living or recently deceased roots have been found in dune soils, except in the Early Holocene dune sands of the Belaya River Valley (Upper Angara Region) of Russia (Golubtsov et al., 2019). In the present study, we characterized pristine rhizoliths in dune soils of the Tengeri Desert (Northwest China), which are formed around deceased roots of *Artemisia*. The factors controlling their formation mechanisms were investigated for the first time in relationship with botanical, pedological and geological parameters. This study provides an excellent opportunity to better constrain the mechanisms of ancient rhizolith formation in dune soils, especially the nature of the interactions between sand minerals and roots in such settings.

2. Geographical setting of the study site

The investigated rhizoliths were found in dune surface soil overlying the bed of the Salt Lake Baijian in the Minqin Basin, central Tengeri Desert, NW China (Fig.1a). Previous studies (Mischke et al., 2016) indicated that a Quaternary mega-lake, Zhuyezhe, was located between the terminal of the Shiyang River and the Laifu and Tangjia Ebo mountains. The present Salt Lake Baijian is the remnant of the Zhuyezhe paleolake (Fig.1b; Chen et al. 1999). Due to the diversion of river water from the upper reach for irrigation and other purposes, the lake has been dried up since the

1950s. Many parts of the paleolake bed have been buried by aeolian dunes and salty marsh deposits (Fig.1b). The main vegetation around the paleolake currently comprises a shrubby or semi-shrubby xerophytic community (Chang et al., 2007). The annual precipitation (110 mm) is dominated by summer monsoon rainfall, which accounts for most of the annual precipitation, and a cold and dry continental air mass that is prevalent in winter. The mean annual air temperature is 7.8°C and the potential evaporation is over 2,600 mm (Long et al., 2010).

3. Methods and materials

Two types of rhizoliths were found in an area of 100 m x 100 m at the study site (Fig. 1): (i) deflated-out/weathered rhizoliths, out of dune soil (Fig. 2) and (ii) pristine rhizoliths, inside the dune soil profile (Figs. 3, 4, 5, 6, 7). Intensive investigations on rhizoliths, soils and plant roots were done with digging where the pristine rhizoliths had been found. Surface soil (20-40 cm thickness) was removed in the locations where the weathered rhizoliths were found, with the aim of finding intact pristine rhizoliths. Many pits (30-50 cm depth) were dug. Preserved pristine rhizoliths were observed in four locations, where they were generally buried under sand soil of ca. 10-30 cm thickness.

Eighteen specimens among the myriad deflated-out/weathered rhizoliths were collected randomly for lab analyses. We paid special attentions to the rhizoliths with root relicts within them when collecting the samples. Yet, only three rhizoliths with black curly threads or hair-like root relicts were found. Four pristine rhizoliths

were only found and collected, which were air-dried at room temperature before laboratory analyses. Some roots of modern *Artemisia* were also collected for element analysis, and one soil sample of ~20cm depth was collected for analyzing bulk carbonate content.

The selected deflated-out/weathered and pristine rhizoliths were analyzed in China Petroleum University (Qingdao, China) using a Zeiss Scope A1 microscope (Zeiss, Carl Zeiss, Germany) and an FEI Quanta 450 FEG (Frequency Electronics, Inc., USA) scanning electronic microscope (SEM) coupled to energy dispersive X-ray spectrometer (EDS). The samples were impregnated with resin, cut into 4.8 cm × 2.8 cm slices and polished to create transverse or longitudinal thin sections to study their micromorphology under a polarizing microscope. Rhizolith fragments were gold coated for conductivity measurements, and then ultra-microscale observations and chemical compositions of the cement by SEM and EDS were acquired. A mixture of Alizarin red S and potassium ferricyanide was used to distinguish among carbonate minerals under the microscope (Dickson 1965, 1966). One sample of pristine rhizoliths containing roots relicts (#Dong I) and one sample of deflated-out/weathered rhizoliths (#Dong II) were radiocarbon dated by accelerator-mass spectrometry (AMS; Beta Analytical Inc., Miami, Florida, USA). This laboratory uses standard procedures for different types of unknown samples (information can be found online at <https://www.radiocarbon.com/beta-AMS-lab.htm>). The measured ^{14}C ages were calibrated to calendar ages using the IntCal13 curve in Calib 7.1 (Talma & Vogel, 1993; Reimer et al., 2013). The $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values of the dating samples were

measured in the laboratory separately using an isotope ratio mass spectrometer. Eight deflated-out/weathered and pristine rhizolith samples were also analyzed for $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ of carbonate cement using a Thermo Fisher Gasbench II-MAT 253 stable isotope mass spectrometer at the State Key Laboratory of Biogeology and Environmental Geology (China University of Geosciences, Wuhan) and the Institute of Geology (Chinese Academy of Geological Science, China), respectively. The detail procedures were previously reported by Song et al (2013, 2014). In addition, the carbonate contents of the bulk soils were determined in Beta Laboratory (Miami, Florida, USA) using phosphoric acid dissolution.

4. Results

4.1. Field observation: rhizoliths, soil and *Artemisia* roots

4.1.1. Rhizoliths

The deflated-out/weathered rhizoliths were observed abundantly at the surface of the sandy soil in several locations around the site. These crumbly rhizoliths lie horizontally on the ground and are randomly scattered. They present short to long branched tube-like structures, with irregular morphology due to intense erosion and weathering (Fig.2). Cross-section observations show that these rhizoliths have mostly circular shapes, with sizes from several millimeters to centimeters. Most of them are hollow and devoid of plant roots, some others being filled with loose dune sand particles and/or showing the presence of dark fiber roots. Shrubs of *Artemisia sphaerocephala* Krasch are still living sparsely in this site (Fig.2; Sun et

al., 2019b).

At the first location, the pristine rhizolith was thick and branched with two lateral crotches along its taproot (Fig.3). This rhizolith likely originates from the deceased roots of *Artemisia*, consistent with the fact that (i) *Artemisia sphaerocephala* Krasch shrubs live there and that (ii) remains of black plant roots were observed within the rhizolith. The rhizolith, shortly after being exposure to the air and sun, became hard and fragile.

At the second location, a pristine rhizolith, a little harder than the surrounding wet soil, was observed under the surface (Fig.4). After a few minutes of air contact both the rhizolith and the soil became dry and white under the sunshine and the blowing winds. This rhizolith, containing deceased root remains, was not connected to any nearby *Artemisia* plant bodies, either living or deceased.

At the third location, a pristine rhizolith, harder than the ambient soil, was discovered just under the surface at 3-5cm depth (Fig.5). Dense, curly deceased hair roots binding sand particles together have been noticed inside this rhizolith (Fig.6).

At the fourth location, a little harder, fragile, horizontally lying rhizolith was found in superficial loose soil (Fig.7). A thread of main root was visible at the two ends of the pristine rhizolith.

4.1.2. Soils

The paleolake sediments are derived from the upstreams of Shiyang River (Qilian Mountains) and the dunes nearby (Fig.1), and correspond to local in situ lacustrine deposits (Mischke et al., 2016; Long et al., 2010). Small low dunes are

being formed on the dry paleolake bed. From a pedological perspective, the soils correspond to dune sands – leptic regosols (FAO, 2014), formed in desert arid climate. Its texture is loose fine to medium size sands (Fig. 3, 4, 5, 6, 7, 8). The upper soil horizon (ca. 0-20 cm depth) is dry (Fig. 2, 3, 5, 6, 7) and the deeper soil is occasionally moist (Fig. 4, 8). A lamellar crust (cemented sand) occurs in some rare parts of the soil surface (Fig. 8).

4.1.3. Roots of *Artemisia*

Numerous lateral roots, deceased and alive, were found within the dry soil horizon (ca. 10 cm depth) but were absent within deeper wet soil horizons (Fig. 8). Hair-like (Fig. 6) and lateral roots (Figs. 3, 4, 5, 7) were observed within the pristine rhizoliths. They are likely derived from *Artemisia* shrubs considering their external appearance and inner texture (Fig. 8) because the *Artemisia* shrubs are the only species growing around the sampling site (Fig. 2, 3). Nevertheless, the modern living roots of the *Artemisia* do not show any evidence of carbonate accumulation and rhizolith formation.

4.2. Laboratory results

4.2.1. Mineralogical and petrological features of pristine rhizoliths

The thin section of rhizoliths (Fig.9a) shows that the main clastic particles within the carbonate-cemented tubes are quartz, feldspars and lithic fragments. Quartz particles do not show clear evidence of weathering, but feldspar particles exhibit weathered rims, along which bright carbonate precipitates. The lithic fragments are motley-colored with stains and speckles.

Staining confirmed that there are lithogenic carbonate fragments in the clastic particles that the cement is calcite with an aphanitic-crystalline texture (Fig.9b). The lithogenic carbonate occurs as coarse fragments and pedogenic carbonate exhibits commonly precipitates in the silt- and clay-size fractions (Kraimer et al., 2005).

The SEM images show that feldspar particles have partially been dissolved or eroded (Fig.10). The cement, calcium carbonate, doesn't show fiber calcite as a biologically-produced mineral such as calcified filaments and calcified fungal hyphae (Sun et al., 2019a; Khormali et al., 2014). There are pores, cavities and voids in the cement (Fig.11, 12a), which is mainly calcium carbonate (calcite) (Fig.12b).

4.2.2 Radiocarbon ages

The AMS ^{14}C dates of two rhizolith samples (Fig.13a, b; a pristine and a deflated-out/weathered, respectively) and the surrounding soil are listed in Table 1. The deceased root relicts are dated as modern, with fractions of modern carbon-14 ($F^{14}\text{C}$) of 1.0000 ± 0.0050 and 1.0138 ± 0.0038 . The calibrated ages obtained using the CALIBomb program (Hua et al., 2013) are AD 1825 $\pm$ 130 and AD 1956 $\pm$ 1, whereas the co-existing rhizoliths cements yield ages of 6355 and 6905 cal year BP (the error is 90 years; Reimer et al., 2013; Table 1). The age of the bulk soil carbonate is 18,670 $\pm$ 220 years cal BP. While the age of the bulk soil carbonate is 18,670 $\pm$ 220 year calBP.

4.2.3 Carbon and oxygen isotopes

The $\delta^{13}\text{C}$ of the carbonate cement varies from -2.05‰ to -3.27‰ (mean = -2.55‰) within the pristine rhizoliths and from -3.20‰ to -6.80‰ (mean = -5.05‰)

within the deflated-out/weathered rhizoliths (Table 2). The relative contributions of the different carbon sources in rhizoliths were estimated using mixing model based on stable carbon isotopic composition ($\delta^{13}\text{C}$). Results suggested that only 9.40% of carbon from decomposed plant roots and 90.6% of carbon from soil carbonates contributed to the rhizolith formation(Sun et al., 2019b). The $\delta^{18}\text{O}$ of carbonate cement varies from -6.01‰ to -7.40‰ (mean = -6.8‰) within pristine rhizoliths and from -2.32‰ to -9.30‰ (mean = -4.54 and -3.56‰ with and without $\delta^{18}\text{O}$ values of Dong II, respectively) within the deflated-out/weathered rhizoliths. The $\delta^{18}\text{O}$ value of the carbonate cement (-9.30‰) in the sample Dong II is the lowest among deflated-out/weathered rhizoliths (Table 2).

The cluster distribution of $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values of the carbonate cement of rhizoliths gave two scopes (Fig.14): (i) the deflated-out/weathered rhizoliths fall in the same part of the diagram, with $\delta^{13}\text{C}$ values $< -3.5\text{‰}$ and $\delta^{18}\text{O}$ values $> -5\text{‰}$, except for Dong II with root relicts and (ii) the pristine rhizoliths fall in the other part of the diagram.

4.2.4 Carbonate contents of the bulk soil

The carbonate content of the bulk soil is 10.5%. Previous work showed that the average carbonate content in Tenggeri Desert is 2.57% (Wang et al., 2004).

5. Interpretation

5.1. Natural conditions of pristine rhizolith formation

The fresh pristine rhizoliths showed that the carbonate cementing was taking

place but was incomplete within shallow soil, with semi-open/semi-closed redox conditions. Comparatively, the deeper wet-saturated soil was in a closed reduction environment. Only the moist soil environment favored complete rhizolith formation in these leptic regosols.

The deflated-out/weathered rhizoliths and pristine rhizoliths occur only at the places where *Artemisia* bushes are growing, suggesting that carbonatization encrustation are formed when this type of vegetation is present. However, the pristine rhizoliths occur only around the deceased *Artemisia* roots, suggesting that the decomposition of deceased roots likely provides organic acids required for carbonate formation, not the respiration of the living roots. Nevertheless, the deceased and living roots of *Artemisia* certainly have rather different physiological and biochemical mechanisms and products.

5.2. Cementing mineral

Microscopy analyses (Fig 9, 10, 11, 12) showed that the cementing mineral, calcite, is homogenous, implying that calcite precipitation occurred in a stable soil environment around the roots. No petrified root texture was observed inside the rhizoliths. This indicated that syngenetic metasomatism and diagenesis did not occur within the shallow soils, the latter always resulting in fossil formation through permineralization, recrystallization, dissolution and replacement in closed redox environments such as sediments and rocks (Brett & Baird, 1986). The calcite cement of rhizoliths may partly originate from bulk soil carbonates.

5.3. Ages of the rhizoliths

Radiocarbon dating (Table 1) indicates that the root relicts are of modern age, whereas the calcite cements are prehistoric, ranging from 6355 to 6905 cal years BP and the bulk soil carbonate is 18,670 cal years BP. Age discrepancies between root relicts and rhizolith cements suggest that the latter incorporated a significant amount of old carbon originating from primary and secondary carbonate minerals within the clastic debris of the soil matrix (Sun et al., 2019b). Such a significant incorporation also indicates the relatively fast formation rates of rhizoliths in the Tengeri Desert leading to the entrapment of old carbon (Wang et al., 2012; Gocke et al., 2011b; Kuzyakov et al., 2006) instead of complete recrystallization and re-equilibration with respired CO₂ of living roots or the atmosphere. Simple mass balance calculation (Sun et al., 2019b; Kuzyakov et al., 2006) demonstrated that contamination of secondary CaCO₃ of Holocene age with even a small portion of primary CaCO₃ can entail strong overestimation of the true radiocarbon age. Nonetheless, the bulk carbonate content of soil is still sufficiently large to cause the reservoir impact on the radiocarbon ages of the dune sediment (X. Yang et al., 2010).

Therefore, the ages of root relicts suggest that the rhizoliths were formed in a very recent time and that the calcite cement ages are biased. Caution should be taken when ¹⁴C ages are used to decipher strata, climate and environmental changes (Sun et al., 2019b).

5.4. Rhizoliths formation environment inferred by C and O isotopes

The δ¹³C and δ¹⁸O values of pedogenic carbonates in deserts and paleosols are good indices to reconstruct paleoclimate and paleoenvironment (Gocke, 2011b;

Monger et al., 2009). Two distinct morphologies of rhizoliths (Fig. 2, 3, 4, 5, 7) with specific $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ values (Table 2, Fig. 14) suggest the deflated-out/weathered rhizoliths on the one hand and pristine rhizoliths on the other hand have been formed under contrasting environmental conditions. The deflated-out/weathered rhizoliths, which have more enriched $\delta^{18}\text{O}$ values (-4.0 to -2.5 ‰) should be formed in a relatively more evaporative condition, comparing to the pristine rhizoliths with $\delta^{18}\text{O}$ values of -9 to -6 ‰. The deflated-out/weathered rhizoliths hence have gone through high temperatures close to the soil surface (Wright et al., 1996) due to strong solar radiation or during the warmest seasons i.e. shortly after summer monsoonal rains. The deflated-out/weathered rhizoliths should also have been formed over relatively short periods e.g. over one growing season when water was available for organisms' activity and root decomposition. This is also evident due to more depleted $\delta^{13}\text{C}$ values (-7.0 to -3.5 ‰) of the deflated-out/weathered rhizoliths referring to higher incorporation of respired carbon than carbon from the entrapped lithogenic carbonates (Table 2). The size, occurrence and $\delta^{13}\text{C}$ values of the pristine rhizoliths (-3.5 to -2 ‰; Table 2) suggest that they have been formed via a mixture of biogenic and non-biogenic processes at deep soil depths (Verrecchia & Verrecchia, 1994, Verrecchia et al. 1995a). As water is nearly available during whole year in deep soil horizons (Fig. 4, 8, 15), $\delta^{13}\text{C}$ of the pristine rhizoliths should be in equilibrium with isotopic composition of HCO_3^- in soil water. The formation of the rhizoliths likely stops at some points because of severe drought, surface soil erosion and denudation by wind and solar radiation, explaining in turn the enriched $\delta^{13}\text{C}$ values of the

pristine vs. weathered rhizoliths (Table 2). These processes might also have led to the exposition of the deceased roots at the surface and/or the end of the root decay, as it most probably occurred for Dong II specimen (Fig. 2, 13b).

6. Discussion: Carbonatization around the deceased roots of *Artemisia*

6.1. Factors controlling the formation of pristine rhizolith

The presence of *Artemisia* deceased roots and soil moisture are the main factors controlling rhizolith formation. Results showed that the pristine rhizoliths were formed by cementing dune sand particles through CO₂ released from decaying *Artemisia* roots. This triggered mineral weathering and calcite crystallization around the roots acting as nuclei. Thereafter, the rhizoliths were deflated out of the soil and weathered under wind and solar radiation. This mechanism of rhizolith formation can be argued as follows.

6.1.1. Deceased *Artemisia* roots

After intensive comparison of the morphological features of the rhizoliths and those of the modern roots of *Artemisia* and other regional vegetation, it appears that the decayed root relicts have the same characteristics as the dried residues of the modern *Artemisia* roots. The general morphologies of the weathered rhizoliths are also mostly similar to those of modern *Artemisia* roots. Therefore, *Artemisia* roots can be considered as the major host material (i.e. nucleus) for rhizoliths formation. The decay of deceased roots may have triggered rhizolith formation, via release of CO₂

during root degradation and production of both HCO_3^- and H^+ , creating acidic conditions, Ca-silicate weathering, carbonate weathering, and driving precipitation of carbonates (Fa et al., 2016; Wang et al., 2015; Berg & Banwart, 2000). This is consistent with the two conditions necessary for the precipitation of carbonate concretions: supersaturation of dissolved carbonate and a nucleus for carbonate precipitation (Raiswell & Fisher, 2000). However, it is unclear why rhizoliths are only formed around the deceased roots of *Artemisia*, rather than any other kind of vegetation.

6.1.2. Soil moisture

The pristine rhizoliths with sub-branches containing deceased hair-like roots (Fig. 6, 7) were discovered horizontally in relatively moist soil and/or upper shallow dry soil horizon but not in deeper continually wet layers (Fig. 3, 4, 5, 7). Thus, besides the deceased roots, the soil moisture content also plays a key role in rhizolith formation (Alonso-Zarza, 1999; Verrecchia et al., 1995b). Nevertheless, if the moisture is too high in the deep soil, the deceased root will be in a reducing environment without enough air exchange, resulting in closed fermentation producing methane rather than carbon dioxide. On the other hand, if the soil is dry without occasional wetness, the root decay will not take place, or will be too slow to release CO_2 and generate HCO_3^- (Sun et al., 2019a). In our sampling site the 10-30 cm depth horizon is the most favorable one for rhizolith formation because of its occasional wetness (Fig. 3, 4, 5, 7, 8) due to climatic and seasonal precipitation variability.

6.1.3. Other factors influencing rhizolith formation

As discussed above, soil moisture and vegetation type (i.e. *Artemisia* shrubs) are the main factors controlling rhizolith formation. Nevertheless, soil moisture is dependent on other factors, such as soil depth, porosity, permeability, landscape position and climate. Based on field exploration the only water sources in Tengger Desert soil are rainfall and snowfall, as no groundwater traces were observed in the sampling site. The sub-branches of the Shiyang River and the Salt Baijian Lake are fed by the upriver streams from the Qilian Mountains. In addition, the Asian monsoon provides rainfall water to the desert area in summer and the westerlies transport snowfall in winter (Zhao et al., 2008). The rainfall and snowfall accumulate into the swales and depressions among the small dunes. Soil moisture varies along the slopes of the dunes, from the crescent peaks to the beds of the swales and depressions. No *Artemisia* is living in the upper slopes of the dunes where soil moisture is very low or in the beds of lakes because of high salinity. *Artemisia* only lives in the beds of the swales and depressions among the small dunes. Roots and rhizoliths can only be observed there (Fig. 2), showing that the morphology of landforms influences the water catchment of rainfall and snowfall and consequently soil moisture required for rhizolith formation in this area.

6.2. HCO_3^- and Ca^{+2} sources for rhizoliths formation

6.2.1. Carbon sources

There are potentially eight C sources for the formation of rhizolith carbonates in desert soils (Sun et al., 2019b): 1) CO_2 from the respiration of living roots; 2) CO_2 from the decomposition of deceased roots; 3) CO_2 from decay and respiration of

organic matter (other than decaying roots) in soil; 4) dissolved ancient pedogenic carbonates; 5) dissolved old lithogenic carbonates; 6) rainfall with carbonate dust; 7) dissolved inorganic carbon from groundwater; and 8) atmospheric CO₂ penetration.

Since no rhizoliths were formed around living roots, CO₂ respiration from such roots cannot be considered as a viable C source. All rhizoliths were observed around deceased roots. The decomposition of the latter provides some carbons necessary for the rhizolith formation, as during such process a fraction of the organic matter is volatilized as carbon dioxide, while residues are increasingly functionalized, with carboxyl, phenol, or hydroxyl groups (Oste et al., 2002; Peinemann et al., 2005). HCO₃⁻ can also be derived from the dissolved old lithogenic carbonates and ancient pedogenic carbonates (Fig. 9b). The contribution of dissolved air-CO₂ in rainfall (carbonate) dust can be considered as negligible due to extremely low annual precipitation (Zhang et al., 2017; Dong et al., 2004), although after rainfall events CO₂ dissolved in rain is sequestered, at least temporarily, from exchange processes between soil and atmosphere, but it is then quickly released to the atmosphere under drier conditions (Serranoortiz et al., 2010). Last, dissolved inorganic carbon in groundwater is not a likely source of carbon in Tenggeri Desert where groundwater was not detected. Mass balance calculation suggests that degradation of plant roots and soil carbonates are the two main carbon sources of rhizolith cement (Sun et al., 2019b; Jin et al., 2015).

The δ¹⁸O of the pristine rhizoliths was remarkably lower than the one of deflated-out/weathered rhizoliths (Table 2), which may be due to oxygen isotope

(H₂¹⁸O) fractionation during soil water evaporation. Indeed, as the evaporation rate of H₂¹⁸O is lower than that of H₂¹⁶O (Merlivat, 1978), the ¹⁸O can be enriched in the deflated-out/weathered rhizoliths which are exposed to the atmosphere, in contrast with the pristine rhizoliths preserved in a wet environment. In addition, the $\delta^{13}\text{C}$ of the pristine rhizoliths was significantly higher than the one of weathered rhizoliths ($p < 0.05$; Table 2). This suggests that the transformation of pristine to weathered rhizoliths when the former are exposed to atmosphere and wind erosion leads to carbon isotope fractionation processes.

The $\delta^{13}\text{C}$ of pristine rhizoliths ($-3.27\text{‰} \sim -2.05\text{‰}$) is much higher than those of the atmosphere (about -8‰) and modern roots (about -18.9‰ , Table 2). The fact that no rhizoliths were found around living roots at the investigated site implies that ¹³C from atmosphere, living root respiration and secondary metabolic processes (living roots of *Artemisia* can secrete organic acid, Deng, 2016) which are ¹³C-enriched (Bowling et al., 2008) cannot directly participate in the rhizolith formation process. Although mass balance calculation suggests that the lithogenic carbonates and pedogenic carbonates are the main carbon sources (Sun et al., 2019b; Jin et al., 2015) for rhizolith formation, soil CO₂ remains an important and direct source of carbon. When the pristine rhizoliths are transformed into weathered rhizoliths, the water is evaporated resulting in enrichment in ¹⁸O and decomposition of H¹³CO₃⁻. Ultimately, the deflated-out/weathered rhizoliths are ¹³C-depleted and ¹⁸O-enriched compared to pristine rhizoliths.

6.2.2. Ca²⁺ sources

Ca²⁺ originates from the weathering of both primary and secondary minerals (Likens et al., 1998) and has therefore typically been thought to persist or accumulate chiefly in semi-arid to arid environments (Rowley et al., 2018). There are several potential Ca²⁺ sources for CaCO₃ rhizolith cement: 1) authigenic carbonates from in situ chemical weathering of soil primary silicate minerals such as feldspar, mica, amphibole, and pyroxene, favored by acidic conditions in relationship with organic matter decomposition (Monger, 2014; Durand et al., 2010) and enhanced by pressure-solution of crystallizations (Monger & Daugherty, 1991a); 2) dissolution of in situ primary lithogenic carbonate rock debris such as limestone and dolomite; 3) dissolution of ancient pedogenic carbonates after many generations of recyclable carbonatization (Monger et al., 2015); 4) dissolved Ca²⁺ from groundwater; 5) dissolved Ca²⁺ in rainfall and snowfall; 6) decomposition of Ca²⁺-rich organic materials (Ranjbar & Jalali, 2012).

First, the in situ chemical weathering of soil primary silicate minerals can provide authigenic carbonates. Nevertheless silicate weathering in surface soils is expected to be extremely slow (Nyachoti et al., 2017; Serranoortiz et al., 2010) due to both the limited amount of rainfall and the relatively neutral soil water pH, with mean lifetimes of 102 to 107 years for carbonate generation (Serranoortiz et al., 2010). The rates of plagioclase dissolution are expected to be even lower in dryland soils, where < 10% of soil Ca is present in silicate minerals and the dissolution kinetics is further limited by mineral surfaces (Nyachoti et al., 2017). For example, it was suggested that the dissolution of silicate rocks contributes to a low extent only to the overall loading

of Ca in pedogenic carbonates of the US Southwest ([Machette, 1985](#); [Capo & Chadwick, 1999](#)). Our bulk microscopy and SEM observations show that the phenomena of silicate weathering and dissolution within the particles are weak and difficult to be seen, with only bright crystalline colour around the clastic particles ([Figs. 9, 10](#)). Therefore, chemical weathering of silicate might represent a minor source of calcium for rhizolith formation in the Tengeri Desert.

Part of the cement is likely derived from authigenic carbonates. Precipitation of authigenic carbonates in soil is accompanied by the formation of Mg-rich clay minerals after the weathering of silicates ([Diazhernandez et al., 2018](#)). In desert with high temperature and salinity, clay minerals like palygorskite and sepiolite are usually formed, favored by the arid to semi-arid conditions and a calcium and magnesium-rich environment ([Verrecchia & Le Coustumer, 1996](#); [Garcia-Romero & Suárez, 2010](#); [Galán & Pozo, 2011](#)). Nevertheless, no authigenic clay minerals were observed in the thin sections of rhizoliths ([Fig. 9, 10](#)). Hence, it is unlikely that the processes operating today were also taking place in the past when rhizoliths were formed ([Diazhernandez et al., 2018](#)).

Second, limestone and dolostone clastic particles do not show obvious dissolution phenomena based on microscopy observations ([Fig. 9b](#)). It is unclear whether carbonate rocks provided enough calcium for rhizolith cementation.

Third, re-dissolution of pedogenic carbonates is impossible to be observed by microscopy and SEM ([Fig. 9, 12](#)) because bio-weathering processes in arid soils with predominance of calcite precipitation are complex ([Monger et al., 2015](#)).

Fourth, no groundwater was found at the sampling site, implying that groundwater dissolved carbonate is not a likely source of Ca^{2+} .

Fifth, rainfall and snowfall are formed through condensation of cloud fresh water vapor or by evaporation of ocean and land water. They do not contain large amounts of salt and cannot provide large amounts of Ca^{2+} , except within rare cases of dust storm rain of carbonate powders (Duan et al., 2007).

The last potential source of calcium is related to the oxalate-carbonate pathway taking place during root decomposition (Verrecchia, 1990). *Artemisia* is among plant species with high oxalate content ($13.40 \text{ g kg}^{-1} \text{ dry matter}$; Huang et al., 2015). Hence, the release of oxalate during root decomposition might be a first step towards carbonate precipitation (Verrecchia 1990; Verrecchia et al., 2006). Altogether, our results suggest that in the Tenggeri Desert, rhizolith formation is primarily connected with root decomposition rather than water uptake by living root. This should be confirmed by further field investigations and lab analyses of roots from different types of vegetation.

6.3. The roles of other chemicals during root decomposition

Besides the acidity produces by CO_2 from decomposition of *Artemisia* roots, other minor chemical compositions of the root are S, N, P, etc, which produce acidic water with the negative ions of SO_4^{2-} , NO_3^- , PO_4^{2-} , etc (Fig. 15). They could enhance and accelerate the weathering of silicate minerals and the dissolution of the primary and secondary carbonate minerals in the soil horizon (Heydari & Wade, 2002; Z. Yang et al., 2010), where abundant carbonate minerals in soil were also found

(Wang, 2016). The carbon dioxide with high gas pressures certainly produced much acidity leading to much carbonate fragments of soil to be dissolved. Indeed, the CaCO_3 solubility in pure H_2O at 25 °C is 0.013 g/L, whereas in weakly acid conditions (e.g. in presence of carbonic acid), the solubility increases up to five times (Aylward, 2007). The function of the inorganic acids above will cause a lower pH soil environment and further enhance particles dissolution and transformation, for instance from feldspar to carbonate. The salts formed by SO_4^{2-} , NO_3^- , and PO_4^{2-} with Na^+ and K^+ can be leached away and deposited in the lowest ponds and lakes of the desert (Yang et al., 2004; Buggle et al., 2011; Liu et al., 2014), as we observed in the Tengeri Desert. Therefore, the relationship between the chemical weathering of silicate minerals and CO_2 released by root decay is not straightforward as acidic conditions may be induced by microbial organic acids and sulfuric and nitric acids (Karim & Veizer, 2000; Spence & Telmer, 2005) derived from the decomposition of *Artemisia* roots (Fig. 15) in addition to CO_2 .

6.4. Redox environment around *Artemisia* roots in soil

The main factors controlling the dissolution and precipitation of CaCO_3 (Gocke et al., 2011a; Drever, 1982) in soil are: 1) CO_2 partial pressure in pore space; 2) pH of soil solution; 3) temperature and 4) mass flow of dissolved carbon species (H_2CO_3 , HCO_3^-). Soil CO_2 partial pressure depends on root respiration and decomposition of organic compounds which subsequently affects soil pH. At the same time, temperature impacts root decomposition rates to produce CO_2 and influences the above mentioned factors (Schlesinger 2016), including the mass flow of acids (H_2CO_3 ,

HCO₃).

The impact of these different factors vary between the soil horizons, with the formation of open, semi-closed or closed systems with respect to soil gas exchange with the atmosphere (Fig 15; Deines et al., 1974). As previously discussed, soil water content and gas exchanges with the atmosphere are vital factors for root decomposition and then carbonate formation in dune soils (Fig. 15). In the superficial horizon corresponding to an open O₂-CO₂ system, desert soils have unlimited supply of dissolved inorganic carbon from the atmosphere and are characterized by low organic matter content due to sparse vegetation. This in turn influences the reactivity and solubility of carbonate minerals (Szramek et al., 2007; Williams et al., 2007).

The rate of root decomposition is high at depths where water content is optimal (Nizami et al., 2018), optimal means water content is most favorable to form a weak redox soil environment without less or too much water. This controls in turn the redox conditions (oxygen content from air) of the rhizosphere and the mineral phase formation (Fig. 15). Soil water fluctuations are controlled by rainfall, water vapor condensation and vertical movement of groundwater. Seasonal climate or diurnal weather changes, including increasing air temperature, evaporation intensity, and faster wind speed, may cause a water vapor deficit across the dunes. Some landscapes, such as interdunal swales, are characterized by optimal moisture conditions for carbonate precipitation by evaporation and rhizolith formation compared to the summits and slopes of dunes or interdunal lakes and ponds. The variable soil moisture conditions across the Tenggeri Desert likely explains why rhizoliths were only found in some, but not all, places around *Artemisia* shrubs.

6.5. Carbonate epidiagenesis in semi-closed desert soil

Rhizoliths form only in semi-wet shallow desert soils, when Ca^{2+} and HCO_3^- become supersaturated around deceased roots of *Artemisia*. This corresponds to a dewatering (evaporative) phase (Breecker et al, 2009), concomitant with a decrease of pCO_2 (Robbins, 1985; Salomons & Mook, 1976). During this transition phase from wet to dry conditions, as Ca^{2+} cations are generally sufficiently abundant in the dune soils, they form carbonate minerals with HCO_3^- , which is confirmed by the carbonate content of the soils. Furthermore, during the decomposition of the roots, the decrease in soil water pH leads to the geochemical weathering of feldspars and other rock debris (Fig. 15) and in turn to the release of Ca^{2+} and other alkali earth metal cations, such as K^+ , Na^+ , Ca^{2+} and Mg^{2+} . As salts based on K^+ and Na^+ are highly soluble in water, they can be leached out and transferred to the ponds and lakes in the desert area. In contrast, Mg^{2+} and Ca^{2+} cations mostly remain in situ. Thus, geochemical weathering continuously and sustainably provides Ca^{2+} cations for the formation of CaCO_3 in arid desert. Laboratory and field studies showed that only weeks or months are needed for fresh, undersaturated waters to reach equilibrium of calcite dissolution, the duration of the process depending mainly on pH and temperature conditions (Serranoortiz, et al., 2010). The transformations through time of clastic particles and soil carbonate minerals (i.e. diagenesis) will likely affect rhizolith formation (Rossi et al., 2001; Mansour et al., 2014). This process should be accurately called epidiagenesis because it takes place in shallow loose sandy soil, without high overlying strata pressure and in connection with the atmosphere (Fig. 2, 3, 4, 5, 6, 7, 8; Table 1).

The cement texture is micritic-aphanitic (Fig 9-12). In general, micrite crystals of calcite indicate initial crystalline stages (Zhou & Chafetz, 2009) and rapid precipitation (Alonso et al., 2004). This is consistent with the environmental conditions in desert, with a quick alternation of day-night temperatures and short shower events. Therefore, strongly alternating wet and dry conditions promote dissolution and precipitation of CaCO_3 and consequently formation of secondary carbonates (Becze-Deák et al., 1997; Borchardt & Lienkaemper, 1999) in desert. In the $\text{CaCO}_3\text{--CO}_2\text{--HCO}_3^-$ system, at higher temperatures, the low solubility of CO_2 decreases the CaCO_3 solubility, and thus promotes CaCO_3 precipitation (Arkley, 1963). On the other hand, a large amount of CaCO_3 has to be in the dissolved state before precipitation, which can be promoted by increasing amounts of dissolved CO_2 from the decomposition of the deceased roots of *Artemisia*. During the day temperatures in the desert are high due to solar radiation on the soil surface without vegetation cover. In these conditions, formation and recrystallization of secondary CaCO_3 require less time ($<10^2\text{--}10^3$ years) than at lower temperatures (Gocke & Kuzyakov, 2011). These processes occur in shorter periods of time if secondary CaCO_3 concretions form around the roots, preventing further dissolution and reprecipitation of CaCO_3 .

The cement, calcium carbonate, does not contain any biologically-produced fibers such as calcified filaments and calcified fungal hyphae (Sun et al., 2019a; Khormali et al., 2014), which might be caused fast by the high temperature in desert.

The higher calcium content of the cement compared to the soil matrix can be

explained by the dissolution of grains due to lithostatic pressure, which is restricted to grain-to-grain contacts (Robins et al., 2012). The force of crystallization which is caused by the growth of secondary minerals subsequently increases the solubility of interacting grains (Monger & Daugherty, 1991a; Robins et al., 2012). The pressure is also generated by the precipitation of calcite crystals that grow and push against silicate grains (Monger & Daugherty, 1991b). In this case, silicate grains do not connect with each other within the calcite matrix, with mainly a basal cement pattern. However, only a vague dissolution of silicate grains occurred at the calcite-silicate connection, with silicate grains matching the shapes of calcite crystals impacted against them (Fig 9, 10; Maliva & Siever, 1988). As a result, pressure-solution might have provided part of Ca source for the carbonate cement formation. Low pH caused by root decay of *Artemisia* (Fig. 15) might be operating in combination with pressure-solution (Monger & Daugherty, 1991a). Altogether, these two processes certainly accelerate chemical weathering of clastic particles and represent major Ca sources. The kinetics of cement formation is also related to the large specific surface area of minerals in sand dunes (Whitfield & Reid, 2013; White et al., 2017), which can enhance chemical reactions (Jung & Navarre-Sitchler, 2018). Mineral-water interfacial area controls the rates of many heterogeneous reactions (Brantley et al., 1999). For instance, weathering rates of feldspars are proportional to their exposed surface areas (Holdren & Speyer, 1985). Reaction rates are also proportional to the number of surface complexes with H, OH, or ligands (Bloom & Nater, 1991). The mechanisms of feldspar dissolution and weathering are still under discussion (Yuan

et al., 2019). In the present study, decomposition of *Artemisia* roots led to more acidic pH conditions and to feldspar transformation to other minerals such as carbonates.

In conclusion, the formation of rhizolith cement in semi-open desert soil is jointly favored by the acidic conditions induced by the decomposition of *Artemisia* root, the pressure-solution of silicate particles, and the large specific surface area of clastic fragments of soil.

7. Conclusions

Well-preserved pristine rhizoliths were found only along the deceased *Artemisia* roots in shallow sandy soil horizons in the swales and depressions among small dunes, of the Tengeri Desert, Northwest China. Rhizolith formation occurred in soils with intermediate moisture, high porosity and permeability, and semi-closed redox conditions in burial depths. These favorable factors firstly facilitated the production of CO₂ from root decomposition where dissolved CO₂ in the soil solution decreased the pH and triggered dissolution of primary and secondary rock debris as well as carbonate minerals. Thus, the decay of deceased roots and minerals weathering provided the carbon sources for the cement formation of pristine rhizoliths. Further, weathering of minerals like feldspars with the influence of pressure-solution and specific surface area of clastic mineral particles were involved in Ca release for carbonate formation. Soil texture and its associated permeability, rainfall fluctuations and its effect on soil moisture content, as well as wind speed variability and dune surface denudation played roles in locations where rhizoliths are formed. The ages of root relicts within the rhizoliths indicated that latter were formed over a relatively

short period of time in the recent decades. The age discrepancies between the carbonate rhizolith tubes and the root fabric relicts illustrated that caution should be taken when interpreting related data and discussing the significance of the rhizolith formation on the evolution of paleogeography, paleoclimate changes, past ecosystems and hydrological variations, as well as when characterizing paleosol development scales. Finally, it remains unknown why rhizoliths are absent around the roots of plant species other than *Artemisia* in the studied area. Further studies are needed to answer this question and especially investigating the biochemical composition of *Artemisia* deceased roots and microbial diversity in the rhizosphere (Liu et al., 2020; Tang et al., 2020).

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984 **Figures and captions**

986 **Fig. 1a.** Location of the study area in Tengri Desert. **Fig. 1b.** Sampling
987 site (red dots) in the Paleolake Zhuyezhe.
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Fig. 2. Fragments of whitish, tubular rhizoliths deflated/weathered on the sandy soil surface of a low dune. The shrub in the background is *Artemisia sphaerocephala*

997 Krasch (the length of the ruler is 20cm). [Fig.3](#). A well-preserved pristine rhizolith
998 were observed after removing the surface sand layer. The small shrubs are *Artemisia*
999 *sphaerocephala* Krasch (the length of the ruler besides the pristine rhizolith is 20cm.).
1000 [Fig.4](#). Moist pristine rhizolith in a wet soil matrix at about 5 cm depth (the length of
1001 the ball-pen is 16 cm). [Fig.5](#). Soft pristine rhizolith within dry sandy soil at about
1002 3-5 cm depth. Living *Artemisia* roots were observed in the vicinity of the rhizolith
1003 (the ruler is 20 cm long). [Fig.6](#). Deceased curly fine hair roots within the pristine
1004 rhizolith shown in Fig 5. [Fig.7](#). A hard pristine rhizolith with a thread of deceased
1005 root in superficial dry loose soil. The bushes are *Artemisia* (the ruler is 20 cm long).
1006 [Fig.8](#). Superficial crust, rhizosphere and moist soil. Beige crust (~5mm thick) is
1007 observed at the surface. A dry loose soil horizon is observed below the surficial
1008 horizon (0.5-25cm deep), with dense fine hair roots showing the rhizosphere. No roots
1009 were found in the moist soil (>25cm depth). The pit depth is ~50cm.

Fig.9a. Microscope image of a rhizolith showing clastic particles and cement. Generally, the particles are not connected with each other mostly inside the cement. The cement has an aphanitic crystalline texture and is homogenous. Along the edges of the particles, bright crystalline carbonate was formed (F-Feldspar, Q-Quartz, D-Rock debris, C-Carbonate cement, P-Pore. Cross polarized light). **Fig.9b.** Microscope image of rhizoliths stained by Alizarin red S and potassium ferricyanide.

The cement among particles was homogenous and stained dark purple red and the two lithogenic-carbonate fragments were stained bright dark purple red (Plane polarized light). [Fig.10](#). SEM image showing feldspar (F) and evidence of dissolution (red arrow) as well calcite cement (C). [Fig.11](#). SEM image of cement, showing tiny pores, cavities and voids among aphanitic and microcrystalline calcium carbonate. [Fig.12a](#). SEM and EDS images of cement, showing aphanitic crystals with tiny pores and voids. Dissolution phenomena are evident in the cement. [Fig.12b](#). EDS spectrum reveals that the cement is made of calcite and low-magnesium calcite.

Fig.13a. Pristine rhizolith (#Dong I, cut from the rhizolith shown in Fig. 3) with hair root relicts selected for AMS radiocarbon dating analysis. **Fig.13b.** Deflated-out/weathered rhizolith (#Dong II; the diameter of the coin is 22 mm) with hair root relicts selected for AMS radiocarbon dating analysis..

Fig. 14. Cross-plot cluster diagram of $\delta^{13}C$ and $\delta^{18}O$ values of the carbonate cement of rhizoliths. Dong II was found at the surface i.e. among the deflated-out/weathered rhizoliths but shows isotopic composition similar to pristine rhizoliths.

Fig.15. Conceptual diagram showing pristine rhizolith formation. Rhizoliths are formed at shallow depths. At such depths semi-closed conditions with moist soil are present. They facilitate root decomposition and carbonate precipitation due to alternation of wet/dry cycles and weak redox conditions. The upper layer is the driest with low amount of water (open oxidation condition), in contrast with sub-layer which is always wet (closed reduction conditions) prohibiting oxidative decomposition of roots.

1072 Tables and captions

1073 **Table 1.** AMS ^{14}C ages of carbonate cement and root hair relicts in rhizoliths

Sample #	Beta lab#	Material	^{14}C yr BP	$\pm (1\sigma)$	Cal age	$\pm (2\sigma)$	F^{14}C	$\pm (1\sigma)$
Dong II	455809	Carbonate	6050	30	6905 BP	90	0.4709	0.0018
Dong II	455808	Root	Modern	---	AD 1956	1	1.0138	0.0038
Dong I	455807	Carbonate	5570	30	6355 BP	50	0.4999	0.0019
Dong I	455806	Root	Modern	---	AD 1825	130	1.0000	0.0050
Soil	514596	Carbonate	15400	40	18670BP	220	0.1470	0.0007

1074 BP: Before present, defined as before 1950; Cal: Calibrated using CALIB7.1 for BP and CALIBomb for AD age.

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1076 **Table 2.** $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values of the carbonate cement of rhizoliths

Rhizolith	Sample No	Material	$\delta^{13}\text{C}$ V-PDB‰	$\delta^{18}\text{O}$ V-PDB‰	Occurrence state of rhizoliths	Lab
Pristine	DH-1	Carbonate	-2.05	-6.01	Within sand soil.	State Key Laboratory of Biogeology and Environmental Geology, China University of Geosciences(Wuhan)
	DH-2	Carbonate	-3.02	-7.30		
	DH-3	Carbonate	-3.27	-6.97		
	DH-A	Carbonate	-2.33	-6.67		
	Dong-I	Carbonate	-2.10	-7.40		
		roots	-18.9			
Deflated-out /weathered	Dong-II	Carbonate	-3.20	-9.30	Newly deflated-out/weathered at soil surface, with root relicts inside the rhizolith.	Beta, Florida, US
		roots	-26.0			
	DH-G	Carbonate	-6.80	-3.62	Deflated-out/weathered at soil surface, without root relicts inside the rhizoliths, at the soil surface.	State Key Laboratory of Biogeology and Environmental Geology, China University of Geosciences (Wuhan)
	DH-H	Carbonate	-6.59	-3.46		
	DH-I	Carbonate	-3.56	-4.02		
	DH-K	Carbonate	-5.11	-2.32		

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V-PDB: Vienna-Pee Dee Belemnite.

- Pristine carbonate rhizoliths with root relicts were first found in Tengger Desert
- Pristine rhizoliths are only forming around the deceased roots of *Artemisia*
- The rhizolith was mainly affected by soil moisture in semi-closed redox soil
- Detail carbonatization of rhizoliths was discussed
- A conceptual model of rhizolith formation was proposed