



Lateral and depth variation of loess organic matter overprint related to rhizoliths – Revealed by lipid molecular proxies and X-ray tomography

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ABSTRACT

Terrestrial sediments like loess are well known for their paleoenvironmental significance. Although organic carbon contents are commonly very low, loess and organic matter (LOM) thereof is regarded as important terrestrial archive for vegetation and climate during deposition. However, the LOM signal is prone to contamination by OM of other age and origin than the synsedimentary vegetation, e.g. by postsedimentary deep-rooting plants. We hypothesized that the influence of rhizosphere effects related to deep-rooting plants varies with depth in quality, quantity and distance to the former root.

The 13 m thick late Pleistocene loess-paleosol sequence at Nussloch (SW Germany) contains rhizoliths (calcified roots) of Holocene age. With the carbonatic encrustation leading to preservation of former root deposits, rhizoliths allow for assessment of rhizosphere processes that occurred during the root's lifetime. Several horizontal transects comprising rhizoliths, surrounding loess (rhizoloess) up to a distance of 11 cm from rhizoliths, and root-free reference loess from different depth intervals, were analyzed for their carbon (C), alkane and fatty acid (FA) composition.

Alkane proxies like carbon preference index (CPI), average chain length (ACL), as well as $n\text{-C}_{27}/n\text{-C}_{31}$ and $n\text{-C}_{29}/n\text{-C}_{31}$ ratios indicated grass vegetation as origin of LOM, while rhizoliths derived from woody vegetation. Several lipid molecular proxies, e.g. short chain/long chain alkanes, long chain alkane composition and long chain/very long chain FA, indicated the incorporation of considerable amounts of root and rhizomicrobial OM. The rhizosphere effect, i.e. postsedimentary overprint of initial LOM in the vicinity of roots, was not restricted to few mm around the former root, but notable to distances of at least 5 cm and possibly more. This was confirmed by X-ray tomography analyses enabling identification of small calcified root remains and non-calcified root channels in the former rhizosphere, which could not be identified under field conditions. In depth intervals with high rhizolith frequency, this entailed lateral overprint of large parts of loess, which in depths of 1.95 and 3.2 m below present surface can cover the whole loess due to extension of the former rhizosphere of >5 cm.

Regarding the high rhizolith frequency (maximizing at 200 m^{-2} at ~2.6 m depth) at Nussloch, these findings raise the importance of deep-rooting plants as potential source for significant postsedimentary overprint of loess-paleosol sequences.

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1. Introduction

Among terrestrial sedimentary archives, loess-paleosol sequences represent one of the most important records displaying Quaternary climatic and environmental changes (Pye and Sherwin, 1999). Bulk organic carbon (C_{org}) as well as individual compound classes thereof (e.g. sugars or lipids) are major tools for studying paleoenvironmental changes throughout loess-paleosol sequences and have been applied,

partly combined with stable isotopes, in many loess settings in Europe and Asia (e.g. Miao et al., 2007; Xie et al., 2003). Such investigations comprise changes in paleoprecipitation, estimated based on bulk C_{org} isotopic composition ($\delta^{13}C_{\text{org}}$; Hatté and Guiot, 2005), and paleotemperature, which is recorded e.g. in archaeal membrane lipids (GDGTs; Peterse et al., 2011). More frequently the paleovegetation was assessed using distribution patterns of alkanes, fatty acids or alcohols (Bai et al., 2009; Zech et al., 2009; Zhang et al., 2006). Such lipidic components can have potential to be preserved in soils and sediments similarly like bulk organic matter (Marschner et al., 2008), justifying their chemotaxonomic and diagnostic character (Maffei, 1996a, 1996b) for reconstruction of paleovegetation and paleoenvironment

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in terrestrial settings. The underlying assumption for these studies is that loess organic matter (LOM) derives from the vegetation present during sedimentation and was incorporated solely during loess deposition, thus representing the environmental conditions during that time. Commonly, the vegetation in loess depositional areas is assumed to comprise mainly scarce grass and herb cover (Liu et al., 1996; Sun et al., 1997), with local occurrences of shrubs or small trees. This has been confirmed by many studies based on lipid molecular proxies (e.g. Bai et al., 2009; Zhang et al., 2006). The second assumption is that LOM is regarded to originate mainly from litterfall of leaves and e.g. rasping of epicuticular waxes from leaf surfaces (Conte et al., 2003).

These presumptions do not take into account that terrestrial settings are exposed also to external sources of organic matter (OM) like e.g. inherited OM from the dust source area (Liu et al., 2007). Also, percolating soil solution was discussed to influence the composition of sedimentary OM (Zhou et al., 1997). Furthermore, plant remains are incorporated into the soil and sediment not solely by litterfall or deposition of abraded particulate OM, but can be derived to a considerable part from roots and rhizosphere processes (Gocke et al., 2010; Jones et al., 2009; Kögel-Knabner, 2002). This has been intensely discussed for the top soil and deep subsoil (i.e. up to 1 m depth; Rumpel and Kögel-Knabner, 2011), but has been frequently ignored for deeper parts of the subsoil and underlying sediments so far, although deep-rooting plants can penetrate sediments up to a depth of several meters (Canadell et al., 1996). However, recent findings strongly support the hypothesis that considerable portions of OM in sequences of terrestrial sediments can be incorporated via roots or rhizodeposits (Gocke et al., 2010; Jansen et al., 2007). The problem of root overprint might exist for any type of terrestrial setting, which is used for paleoenvironmental reconstruction. In loess, the risk of considerable alteration of the original signal by postsedimentary deep-rooting plants is particularly high because of very low C_{org} contents (commonly $\leq 3 \text{ mg g}^{-1}$), making loess prone to contamination by external sources of OM (Head et al., 1989; Zhou et al., 2005). While LOM is not necessarily altered in quantity, its composition can be considerably changed in vicinity of roots due to incorporation of root and rhizomicrobial remains and rhizomicrobial re-working of LOM. This may have also a strong effect on paleoenvironmental reconstructions based on LOM.

Highest rhizomicrobial activity and thus largest amounts of rhizodeposits occur commonly around fine root hairs. With an average longevity of approximately one year (Strand et al., 2008) and turnover of few years (Fröberg, 2012), these fine root hairs may not be visible anymore after degradation and thus difficult to study, except if roots were surrounded by precipitates of e.g. iron oxides, silicon oxides or carbonates (Cramer and Hawkins, 2009; Gocke et al., 2010; Jones et al., 1998). This process, leading to formation of rhizoliths, is likely to take place during the root's lifespan or shortly thereafter (Gocke et al., 2011; Jones et al., 1998), thus resulting in preservation of former root tissue and microbial remains due to encrustation by minerals (Gocke et al., 2010).

Although it is obvious that roots alter the chemical composition in their direct vicinity (few mm) (Sauer et al., 2006), it remains unclear, how far such alterations can be expected at larger distance from the surface of rhizoliths. The main aim of the current study was to identify the extension of the former rhizosphere around rhizoliths and thus assess the significance of such postsedimentary processes for studies focussing on LOM. The study covers rhizoliths and former rhizosphere taken at different depth intervals of the Nussloch loess-paleosol sequence (Antoine et al., 2001, 2009; Zöller et al., 1988). These rhizoliths consist mainly of CaCO_3 (Gocke et al., 2011). The rhizosphere was sampled up to 10 cm surrounding the rhizoliths. Rhizosphere changes were determined for a variety of molecular proxies covering lipidic fractions of *n*-alkanes and fatty acids. Additionally, for the first time intact sediment cores from loess surrounding rhizoliths were amended to X-ray tomography (CT) to visualize remains of the former root systems without destructive sampling. This methodology has high potential for the study of biopores generated by root systems in soil (e.g. Gregory et al., 2003;

Tracy et al., 2010), but was to the best of our knowledge never tested on rhizoliths developed in terrestrial sediments.

We hypothesized that rhizoliths at Nussloch derived from different plants in different depths of the loess-paleosol sequence, therefore influencing their surrounding in various ways concerning accumulation and/or depletion of C_{org} and individual compound classes. We further hypothesized that the influence of rhizosphere processes, i.e. the alteration of original LOM, can strongly vary with depth in lateral dimension and intensity.

2. Material and methods

2.1. Study site

The Nussloch loess-paleosol sequence is situated in Central Europe near Heidelberg, SW Germany (49° 18.724' N, 8° 43.381' E, 217 m a.s.l.). Its stratigraphy, sedimentology and paleopedology have been intensely investigated since several decades (Antoine et al., 2001, 2009; Lang et al., 2003; Zöller et al., 1988). In the still active open castmine of the HeidelbergCement AG, the last glacial cycle, comprising the last 130 kyrs, has been studied mainly in four standard profiles (P1–P4), which were described in detail by Antoine et al. (2001, 2009).

Samples for this study were collected between April and July 2011 at a slope, 12 m N of the profile described by Gocke et al. (2011a) and about 600 m WSW from the standard profile P4 (Antoine et al., 2001). The area was used for agriculture until 2004 and left fallow until sampling, thus enabling primary vegetation to invade on the top soil and parts of the slope, which was generated in 2006 to access the underlying Upper Muschelkalk (Middle Triassic) limestone for mining. The slope extends several tens of meters from NW to SE.

The sampled profile comprised 13.1 m of loess and soil units including the recent soil as well as three main paleosols and 14 incipient paleosols. Carbonate concretions including loess dolls and rhizoliths were abundant (Gocke et al., 2011).

2.2. Sampling and sample preparation

The profile was produced by removing at least 1 m of material from any side (i.e. top and side of the sampled interval) to reduce contamination of LOM (e.g. by percolating soil solution, living roots penetrating the slope, and infiltrated precipitation) to a minimum. To count rhizoliths in individual depths, planar levels with approximate dimensions of 1.4 m length and 0.8 m width were created in the individual sampling depths by removing all sedimentary material above the respective level. After assessing the stratigraphic profile, loess sediments were selected for sampling, which were located in between initial or well developed paleosols, thus allowing to study the influence of rhizosphere processes on C_{org} poor terrestrial sediment without interference by (paleo)pedologic processes. Throughout the profile, the abundance of rhizoliths was determined on each planar level by a grid with a side length of $0.5 \times 0.5 \text{ m}$ and extrapolated to a scale of 1 m^2 . This methodology of counting did not take into account smaller rhizoliths ($< 0.1 \text{ cm}$) or other forms of carbonate precipitates potentially formed around fine roots of grass vegetation, which account for up to 20,000 per 1 m^2 (unpublished data). Their frequency was determined only in few selected depth intervals (not shown here). The depth intervals selected for the current study were free of such accumulations except for 6.0 m.

In the respective depth intervals (1.95 m, 3.2 m, 4.2 m, 5.4 m, 6.0 m, 7.4 m and 13.0 m), rhizoliths (R) were selected for sampling, which revealed no or low numbers of visible root remains in the surrounding, the so-called rhizoloess (RL). Especially in the upper part of the profile with rhizolith occurrence of $> 100 \text{ m}^{-2}$ other root remains in the surrounding could not be avoided and therefore large rhizoliths (diameter $> 1 \text{ cm}$) were selected, where other root remains were much smaller ($< 0.2 \text{ cm}$). Plastic rings, each with an inner diameter of 20 cm and a

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