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New frontiers in the molecular based reconstruction of Quaternary paleovegetation from loess and paleosols

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ABSTRACT

The reconstruction of paleovegetation from terrestrial sediments such as loess and paleosols represents a major, but still challenging task in Quaternary research. A significant methodological advance was the implementation of biomarker applications to terrestrial archives such as loess sequences and fossil soils. Essentially, the n-alkane approach has been widely applied within the last years to reconstruct changes in the paleoenvironment as for example regarding the relative abundance of woody species versus grasses from Quaternary terrestrial deposits. However, complex distribution patterns of leaf wax lipid homologues in plants and often little taxonomic specifity limit the use of n-alkyl series in paleoflora research. In this study, retene is introduced as a new molecular marker specifically for conifer derived organic matter to Quaternary loess – paleosol research. The ratio of retene to cadalene serves as a useful proxy for the relative contribution of conifers to the paleovegetation. Up to now these molecular markers have been mainly applied to coal research and marine sediments. Using samples from the Tumara site (Siberia), the added value of this new molecular approach for paleovegetation reconstruction also for loess and Quaternary fossil soils is demonstrated. The results of this pilot study confirm the presence of conifers during the penultimate glacial in NE-Siberia.

1. Introduction

1.1. Approaches for paleovegetation reconstruction from Quaternary terrestrial archives

Loess deposits and therein buried fossil soils represent the most widely spread terrestrial archive of the Quaternary paleoenvironment. Loess and fossil soils have been intensively investigated for paleoclimatic proxies in various regions of the world. However, knowledge on Quaternary paleovegetation derived from these archives is still scarce. This is mainly due to the inadequate implementation of paleofloristic proxies in Quaternary terrestrial sediments and fossil soils. Plant macrofossils are among the most valuable vegetation proxies. They provide distinct evidence for plant species in the local paleoenvironment on high taxonomic level, but as plant macro-remains are rarely fossilized in fossil soils and terrestrial sediments such as loess, they give only a discontinuous picture on the paleovegetation. Hence, plant macrofossils often represent only a snapshot proofing the presence of single species at a short time interval of the past (Rudner and Sümegi, 2001).

Another type of evidence on past vegetation changes can be provided by microfossils such as phytoliths, mollusks, or pollen. Pytoliths and snails reflect mainly habitat types and the ecological characteristics of habitats in the close surrounding of the geoarchive (e.g. Sümegi and Krolopp, 2002; Barczi et al., 2009). However, limited information is provided on specific plant taxa and in alkaline environments, phytoliths have only a low preservation potential, while in decalcified sediments and paleosols, mollusks are only rarely preserved (Marković et al., 2007; Sümegi et al., 2011). Pollen analysis allows the identification of plant taxa mostly to family and in certain cases even to species level (Gerasimenko and Rousseau, 2008; Komar et al., 2009). However, the source region of pollen is often less well definable due to far distance transport and especially in oxic sediments palynological records can be biased by selective preservation of palynomorphs. Hence, under-, or overrepresentation of plant taxa in the pollen record is a crucial issue in the interpretation of pollen assemblages in loess and fossil soils (e.g. Faegri and Iversen, 1989; Birks and Birks, 2000; Gerasimenko and Rousseau, 2008).

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In contrast, fossil organic material is generally present in loess as well as in paleosol units, though the concentrations can be as low as few mg TOC per gram loess or fossil soil material. Marker molecules of certain plant taxa preserved in the fossil organic material can be regarded as molecular fossils or biomarkers. Such molecular fossils, being part of the fossil organic material, mainly derive from the on-site or near to on-site paleovegetation, and hence can be used for continuous records of the autochthonous paleovegetation in loess-paleosol sequences.

In recent years, molecular approaches to characterize paleovegetation from Quaternary terrestrial sediments and fossil soils mainly focused on long-chain n-alkyl series, essentially long chain n-alkanes (Zhang et al., 2006; Bai et al., 2009; Zech et al., 2010, 2011a; Gocke et al., 2013). Long-chain n-alkanes mainly derive from cuticular plant waxes. For woody species often a dominance of the homologues n-C27 and n-C29 has been reported, whereas for grasses especially of the family Poaceae a dominance of the homologues n-C31 and/or n-C33 is typical (Cranwell, 1973; Meyers and Ishiwatari, 1993; Maffei, 1996a, 1996b; Zech et al., 2009a, 2009b, 2010). Hence, within recent years long chain n-alkane ratios (LARs) such as n-C27/n- C31 and (n-C27 + n-C29)/(n-C31 + n-C33) have been well established in loess-paleosol research to characterize the relative abundance of woody vs. grassy species (e.g. Zhang et al., 2006; Bai et al., 2009). Within recent years, there has been significant progress in the n-alkane approach, as post-sedimentary alteration effects such as degradation have been studied more closely, ending in the implementation of various corrections procedures (Zech et al., 2009b, 2011b; Buggle et al., 2010). An endmember modeling approach has been established by Zech et al. (2009b) for a rough estimation of the quantitative abundance of woody versus grassy species.

The "classical" n-alkane pattern proposed for the differentiation of grassy and woody types of vegetation is valid for many species in the boreal and temperate biomes of Eurasia, but also numerous exceptions have been reported (Maffei et al., 2004; Diefendorf et al., 2011; Bush and McInerney, 2013; Tarasov et al., 2013), complicating the application of the n-alkane approach for paleovegetation reconstruction. Moreover, in most environmental settings, the chemotaxonomic significance of n-alkanes is not good enough to identify woody plants on a higher taxonomic level e.g. to differentiate between trees and shrubs. In NW China for example, a dominance of n-C27 and n-C29 has been also reported from leaf waxes of Chenopodiaceae shrubs, so that some authors interpret the ratio of n-C27/n-C31 and n-C29/n-C31 in Chinese loess-paleosol sequences in terms of relative abundance of Chenopodiaceae in the paleovegetation (Liu and Huang, 2005). In addition, it has been shown that slight variations of the n-alkane distribution can also occur in the wake of climatic changes (e.g. Dodd and Afzal-Rafii, 2000; Shepherd and Griffiths, 2006). Nevertheless, the n-alkane approach is often regarded as state of the art in molecular-based vegetation studies in loess and Quaternary paleosols, although, in most cases the n-alkane approach is at best restricted to give a rough estimate on the relative contribution of woody species in the paleovegetation. Therefore, it is desirable to complement the n-alkane approach by vegetation fingerprints in other lipid fractions and other types of plant biomarkers. Certain biomarkers, especially of the terpenoid class, provide a potential key to more specific information on the paleoflora composition.

1.2. Principles of terpenoids and terpenoid-derived compounds as molecular fossils

Terpenoids are the main components of tree resin. Some substances in this class of compounds represent chemotaxonomic markers i. e. they are taxon specific so that only from their identification in a fossil soil or sediment can conclusions on the former presence of the respective taxon be drawn (Otto et al., 1997; Otto and Wilde, 2001; Hautevelle et al., 2006). An overview of the most important differences in terpenoid biomarker signature for various tree taxa is presented in Fig. 1. For a more extensive review on terpenoid biomarkers we refer to Otto and Wilde (2001) and Diefendorf et al. (2012). Briefly, triterpenoids of oleanane, ursane and lupane type are characteristic of angiosperm-derived resin, while diterpenoids are almost only produced by gymnosperms. Many degradation products of these terpenoids retain the chemotaxonomic significance. Retene (1-methyl-7-isopropyl phenanthrene) is a common degradation product of conifer-derived abietane-type diterpenoids. Due to its structure (aromatic, no polar functional group) it is assumed to be relatively stable against biodegradation. Therefore, it potentially allows the identification of conifer-derived organic matter even after strong degradation (Laflamme and Hites, 1978; Hautevelle et al., 2006). Cadalene is a degradation product of plant-derived sesquiterpenoids of the cadalene type. These cadalene-type terpenoids are generic i.e. they can be found in all vascular plants (Simoneit, 1985; Hautevelle et al., 2006). As the precursors of retene and cadalene are assumed to have a similar sensitivity for biodegradation and due to structural similarity, Van Aarsen et al. (2000) and Hautevelle et al. (2006) argued that the relative abundance of retene and cadalene in sediments primarily reflects variations in the source of higher plant derived organic matter. Van Aarsen et al. (2000) introduced the ratio of retene to the sum of retene and cadalene as higher plant parameter (HPP) and Hautevelle et al. (2006) implemented the ratio of retene to cadalene as indicator for the contribution of conifers to the paleovegetation. Both ratios have been well established as proxies for detecting source variations of higher plant derived material (e.g. Okano and Sawada, 2008; Izart et al., 2012).

In spite of the high potential of terpenoids and terpenoid-derived biomarkers for reconstructing paleovegetation, this approach, up to now, has almost only been applied in lacustrine and marine sediments, as well as lignites (Otto et al., 1997; González-Vila et al., 2003; Otto et al., 2005; Hautevelle et al., 2006; Bechtel et al., 2007; Okano and Sawada, 2008; Izart et al., 2012). There is no study known showing the feasibility of this approach for paleoflora reconstruction from fossil organic material in loess and Quaternary paleosols. In the following, we report on a pilot study investigating the potential of terpenoid-derived markers for paleovegetation reconstruction from a Quaternary loess-paleosol sequence.

2. Material and methods

2.1. Study site and regional setting

We chose the Tumara site in NE Siberia, (63°36′ N, 12°58′ E) to investigate the potential of retene for paleoflora reconstructions from loess and Quaternary paleosols, because this site has a well documented stratigraphy, microfossils (pollen) and n-alkane biomarker records (e.g. Zech et al., 2008, 2010, 2013; Zech, 2012). This previous work offers the possibility of cross-evaluating the various paleovegetation markers with the retene record. The study site is situated south of the Verkhojansk Mountains at the banks of the Tumara River (Fig. 2). The modern climatic conditions in this area are highly continental with minimum temperatures in the range of −60 to −70 °C and maximum temperatures of +38 °C and a mean annual rainfall between ~155 and 210 mm (Zech et al., 2010 and references therein). Larch forests dominated by *Larix dahurica*, *Betula pendula* and *Betula alba* characterize the present day vegetation in the region. The Tumara site comprises a sequence of

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