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Radiogenic and “stable” strontium isotopes in provenance studies: A review and first results on archaeological wood from shipwrecks

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ABSTRACT

Different approaches are used to study wood provenance, but most of them are based on tracers in wood that are generally controlled by climatic factors. The strontium isotopic ratio $^{87}Sr/^{86}Sr$ in trees and soils is related to the signature of the local bedrock. Despite being used in diverse archaeological studies, Sr isotopes have rarely been used to trace the provenance of archaeological wood and especially wood from shipwrecks. In addition, recent analytical advances have allowed the detection of mass-dependent fractionation of Sr isotopes during biogeochemical processes, as reflected in the variation of $\delta^{88/86}Sr$ values between different environmental materials. The $\delta^{88/86}Sr$ values could be used in conjunction with the $^{87}Sr/^{86}Sr$ isotope ratio to improve constraints on the sources of Sr in the archaeological materials being studied. This paper discusses the potential and limitations of using both of these Sr isotope ratios to trace the provenance of wood from shipwrecks. We review the $^{87}Sr/^{86}Sr$ and $\delta^{88/86}Sr$ variations in rocks, waters, soils, plants and other living organisms and discuss how to determine the local Sr isotopic signature of potential sites. We also compile a list of known wood *post mortem* modifications in seawater. Possible implications in terms of the modification of the original Sr isotope ratios of wood during storage in seawater are illustrated through preliminary observations. This paper points out some limitations and perspectives for using Sr isotopes in provenancing wood from shipwrecks, and suggests future research to test and apply this approach for tracing the origin of archaeological wood.

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1. Introduction

The study of wood provenance (dendroprovenancing) can be applied for many purposes, including identifying the source of the construction wood used for building monuments, ancient houses and ships, understanding wood trading links in the past, and tracking illegal timber logging. Any physiological or chemical tracer of wood provenance must be highly specific of the geographic area of origin. Dendrochronology, wood anatomy, genetics and biogeochemical analyses are among the different tools that can be used to determine the provenance of wood. The wood in question needs to be compared with data on wood (or other biogeochemical compartments) from the potential sites of origin. Therefore, we need a comprehensive and large dataset on the tracer, at spatial scales varying from the region to the catchment or even the stand. However, in the case of archaeological wood, and regardless of the selected technique, some methods should be used with caution and adapted if processes such as diagenesis have modified the wood structure and/or composition during burial in soils or sediments.

Dendrochronology is the dating and study of annual rings in trees, representing the most common method developed so far for wood provenance studies. Trees respond to the climate by adapting their growth rate. As a result, trees growing in a given geographical location and experiencing similar environmental conditions (*e.g.* annual rainfall, air relative humidity, soil moisture, ground water availability) will display similar tree-ring patterns and typical pointer years. Dendrochronology has been used successfully to determine the origin of diverse wood materials *e.g.* wood from shipwrecks (Bonde and Christensen, 1993; Daly, 2007; Daly and Nymoen, 2008; Domínguez-Delmás et al., 2013), wine barrels (Eckstein et al., 1975), furniture and altarpieces (Haneca et al., 2005) as well as wooden foundation piles of historical buildings (Sass-Klaassen et al., 2008). The use of dendrochronology can be complicated in some cases when only short chronologies are available (Billamboz, 2003). Short tree-ring series (less than 60 years) yield lower correlation coefficients when cross-dated and lead to a higher probability of dating errors compared to longer chronologies, hence long tree-ring series are required to ensure a satisfactory interpretation. Another limitation can arise from the presence of various growth anomalies in the studied timber (Haneca et al., 2009). While extensive tree-ring databases are established in some regions (*e.g.* northern and central Europe), more data are needed in other regions to cover wider geographical areas and acquire longer chronologies (Domínguez-Delmás et al., 2015).

Alternatively, wood anatomy can serve to identify plant taxa to the genus level and sometimes to the species level. If the genus or species can be determined, and have small area of distribution linked to environmental conditions such as climate, this method might be suitable for wood provenance studies. In particular, Esteban et al. (2012) showed that the variation of anatomical features linked to provenance and microclimate conditions is greater than the variation within populations of *Pinus nigra* from 17 different regions in Spain. This observation shows the potential of using wood anatomy for provenance studies by matching the anatomical features of the examined wood with material from the site of origin. However, more studies are needed to validate this approach in other areas and with other species.

Genetics provides another tool for identifying the provenance of wood. For example, during the Quaternary glacial episodes, European forests were restricted to the Iberian, Italian, and Balkan peninsulas (Petit et al., 2003). Afterwards, the chloroplast DNA (cpDNA) lineages of European oaks became widely distributed due to post-glacial colonization outward from the different European refugia. This resulted in a clear geographical structure of cpDNA haplotypes (Deguilloux et al., 2004, 2003, Petit et al., 2002a, 2002b, 2002c). By characterizing the cpDNA haplotypes of oaks in Ireland, Kelleher et al. (2004) concluded that they matched the cpDNA of Iberian refugia out of which oaks migrated following the last glaciation. This method has a limitation when haplotypes are distributed over large areas. Moreover, when the wood comes from plantation trees, the DNA fingerprints do not match the natural distribution of DNA (Kagawa and Leavitt, 2010).

The compositions of cellulose, hemicellulose and lignin (Sandak et al., 2011), as well as other wood constituents such as so-called "wood extractives" (Miranda and Pereira, 2002) or monoterpenes (Smith et al., 1969) can be significantly different in relation to their specific geographical distribution. These variations in wood composition between geographical areas are linked to different environmental and climatic factors (Sandak et al., 2015). Clarke et al. (1997) found significant differences between cellulose and pentosan components in different species of *Eucalyptus* from different provenances. Rodrigues et al. (1998) showed variations in lignin content, which make it possible to trace the provenance of *Eucalyptus globulus*. These provenance studies using organic tracers are encouraging insofar as they reveal distinct organic compositions of certain species according to specific areas. However, further studies are needed to establish suitable species and organic component that could be used.

The isotopes of a given element show a shift in their relative abundances during biogeochemical cycling due to biological and physico-chemical processes – this phenomenon is known as "isotope fractionation". In particular, climatic conditions influence the isotopic ratios of hydrogen (H), carbon (C) and oxygen (O), *i.e.* the main elements present in wood. While the signature of C isotopes in plants is linked to soil moisture and depends on relative humidity, temperature and precipitation (Francey and Farquhar, 1982; Saurer et al., 1995; Stuiver and Braziunas, 1987), the H and O isotope ratios are influenced by spatial variations of precipitation and humidity (Barbour, 2007; Bowen et al., 2005; Farquhar et al., 1997; McCarroll and Loader, 2004). As a result, the wood from growth-rings can record a specific isotopic fingerprint linked to the climatic factors at the original site of the tree (Leavitt, 1993). Kagawa et al. (2007) found a significant correlation between