



Seasonal changes in molecular composition of organic matter in lake sediment trap material from Nylandssjön, Sweden



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ABSTRACT

The origin and degradation/preservation state of organic matter (OM) in lacustrine systems can be studied using its molecular composition. Pyrolysis gas chromatography–mass spectrometry (Py–GC–MS) allows rapid assessment of OM-rich samples and has proven a useful tool for peat, lacustrine and marine deposits, but is rarely applied specifically to lake environments prone to the formation of varves (annually laminated sediments), which are used frequently for palaeoenvironmental reconstruction. We applied Py–GC–MS to a 3.5 yr record of sediment trap samples collected from Nylandssjön in northern Sweden to determine the usefulness of the method for improving knowledge of OM dynamics in varved lakes. It appeared that seasonal variation controlled the pyrolysis fingerprints: intact polysaccharides and lignin from vascular plants, chitin from arthropods and chlorophyll from algae were concentrated in spring and summer traps, whereas winter trap samples were enriched mainly in degraded components, as OM slowly trickled down under a thick layer of ice. A secondary diagenetic process involving sulfuration of phytadienes was recognised via isoprenoid thiophenes, with no clear seasonal trend, probably because it occurred under anoxic conditions at the lake bottom. The N-containing compounds from proteins in algae and chitin in arthropods were abundant in all samples, indicating that the dominant flux of varve OM was autochthonous in origin. We conclude that Py–GC–MS has significant potential for identifying the variability in parent material (source organisms and degradation state) and could become a valuable tool for palaeoenvironmental reconstruction from varved lake sediments.

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1. Introduction

Lake sediments in general, and annually laminated (varved) ones in particular, have proved to be useful archives of environmental change on both long and short timescales (Segerström et al., 1984; Schaller et al., 1997; Vos et al., 1997; Brännvall et al., 1999; Moore et al., 2001; Ojala et al., 2008; Petterson et al., 2010; De Jong and Kamenik, 2011). However, to take full advantage of the information recorded in the varves there is a need for a better understanding of the environmental factors which affect their formation (Lotter and Birks, 1997; Gälman et al., 2006).

One of the most intensively studied lakes with varved sediments is Nylandssjön, a small lake in northern Sweden (Fig. 1). The varves have been studied to identify the effect of external environmental forcing (Gälman et al., 2006; Rydberg and Martínez Cortizas, 2014) as well as internal mechanisms of sediment

alteration via post-deposition processes, i.e. to assess the effects of diagenetic change (Gälman et al., 2008, 2009a,b; Rydberg et al., 2008; Boës et al., 2011; Klaminder et al., 2012; Maier et al., 2013). From studies of the concentrations of C and N (Gälman et al., 2008) it was apparent that the effect of diagenesis was different for varves from different years, the differences presumably being due to differences in the composition of the sediment, especially the organic matter (OM) fraction.

The OM in a lacustrine environment can be expected to consist of varying proportions of four main biological sources: (i) terrigenous vascular plants and emergent macrophytes, composed mainly of polysaccharides, lignin, non-lignin phenolic acids, resins, tannin, plant wax and aliphatic biopolymers (cutin, suberin); (ii) vascular aquatic plants (submerged and floating macrophytes), with a similar general composition but with negligible lignin (Thomas et al., 2009); (iii) microphytes (algae, including phytoplankton and cyanobacteria), containing polysaccharides, chlorophyll, protein, phlorotannins and aliphatic biopolymers including algaenan (Gelin et al., 1996); (iv) arthropods (water fleas, chironomids,

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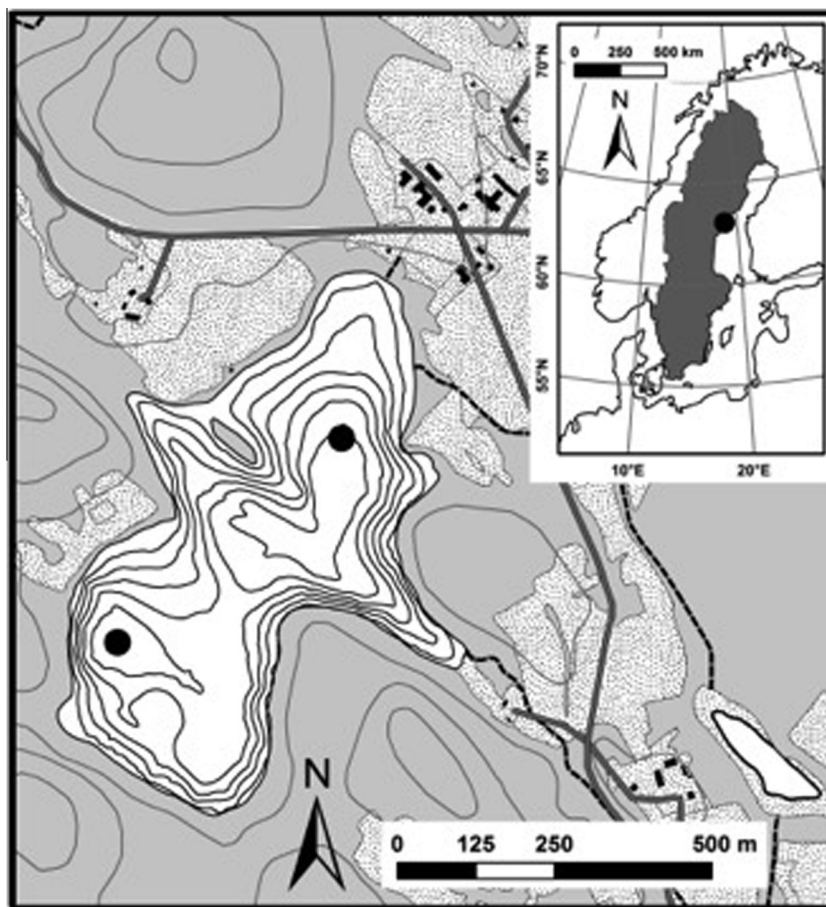


Fig. 1. Location of study site. The bathymetric map insert shows the two depocentres of the lake, with the 17.5 m deep main basin in the northeastern section.

etc.), composed mainly of polysaccharides, including chitin (a polymer of *N*-acetylglucosamine) and proteins (Meyers and Ishiwatari, 1993; Kögel-Knabner, 2002). Both between years and over the course of a single year, the relative importance of these sources will vary and the resulting differences in chemical and physical structure of the OM will probably affect the nature and intensity of diagenetic alteration. To study how the OM composition varies between both seasons and years there is a need for an analytical tool that can provide detailed information on its molecular composition and origin.

Pyrolysis gas chromatography–mass spectrometry (Py–GC–MS) combines thermal degradation under an inert atmosphere (pyrolysis), with chromatographic separation and spectrometric identification of pyrolysis products to assess the molecular composition of intractable material such as synthetic and natural organic polymers (Moldoveanu, 2001; White et al., 2004). It has been applied intensely since the 1980s to OM from soils and sediments (Sáiz-Jiménez, 1988 and references therein) and specific biopolymers such as cellulose (Pouwels et al., 1989), lignin (Faix et al., 1987), proteins (Tsuge and Matsubara, 1985), chitin (Stankiewicz et al., 1996), aliphatic biopolymers (Nip et al., 1986), chlorophyll (Ishiwatari et al., 1991) and tannin (Galletti et al., 1995). It is a convenient method for the rapid assessment of bulk samples from OM-dominated soils and sediments, because it requires minimal or no sample pre-treatment and only a small amount of sample, which is paramount in the analysis of varved lake sediments, where sample availability is often limited. On the other hand, pyrolysis fingerprints of complex mixtures are difficult to interpret because many pyrolysis products have multiple potential

precursors and because of secondary rearrangements and matrix effects (Saiz-Jiménez, 1994), which is why Py–GC–MS has been referred to as a double edged sword that calls for cautious interpretation (Sáiz-Jiménez, 1996). The method is rarely applied to high resolution palaeoenvironmental reconstruction from lacustrine or marine systems, contrary to terrigenous (peat) deposits (e.g. Boon et al., 1986; Buurman et al., 2006; McClymont et al., 2011; Schellekens, 2013). A relevant exception is the contribution of Fuhrmann et al. (2004), who used it to discriminate the main OM sources in a varved sediment in order to estimate the relative proportions of autochthonous (algae) and allochthonous (macrophytes) OM based on the balance between lignin phenols and aliphatic pyrolysis products.

In this study, we have applied Py–GC–MS to samples from sediment traps in Nylandssjön. The main objectives were to (i) build the framework necessary for successful interpretation of pyrolysis fingerprints from OM in varved lake sediments and (ii) increase understanding of varved sediment formation, especially in relation to how OM composition varies between seasons, depending on the input from different OM sources.

2. Materials and methods

2.1. Study site

Nylandssjön (62°57'N, 18°17'E) is a small (0.28 km²), circum-neutral, mesotrophic and dimictic boreal forest lake at the coast of the Gulf of Bothnia in northern Sweden

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