Palynofacies as useful tool to study origins and transfers of particulate organic matter in recent terrestrial environments : synopsis and prospects.

David Sebag, Yoann Copard, Christian Di Giovanni, A. Durand, Benoît B. Laignel, S. Ogier, Elisabeth Lallier-Vergès

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D. SEBAG[a], Y. COPARD[a], Ch. DI-GIOVANNI[b], A. DURAND[a], B. LAIGNEL[a], S. OGIER[a], E. LALLIER-VERGES[b]

[a] *UMR CNRS 6143, M2C, Département de Géologie, Université de Rouen, 76821 Mont-Saint-Aignan Cedex, France,*

[b] *UMR CNRS 6113, ISTO, Bâtiment Géosciences, Université d'Orléans, BP 6759, 45067 Orléans Cedex 2, France*

Corresponding author: E-mail address: david.sebag@univ-rouen.fr

Abstract

Palynofacies analysis is based on transmitted light microscope study of organic constituents isolated and concentrated by acid and basic digestions. Published results of studies of present-day terrestrial environments show that two complementary approaches successfully characterize particulate organic matter (OM) from palynofacies analyses. The first method is based on the identification and the quantification of some typical particles (*optical markers*) according to their origin (i.e. aquatic or terrestrial), their nature (i.e. biogenic, anthropogenic, fossil), and/or their formation (i.e. biodegradation, combustion, oxidation). The second approach is based on the use of binary or ternary diagrams in order to define *petrographical signatures* from the relative proportions of significant organic constituents. This approach can be used for tracking i) changes in OM composition during humification in soil profiles, ii) transport of reworked terrestrial particles, iii) diagenesis of peaty deposits, or iv) weathering of geological substratum. The more advanced approach is based on the use of some predefined optical markers and their optical signatures to establish the relation between the OM compositions (*palynofacies*) and their depositional environments. In addition, this kind of study aims to define a modern frame of reference that can be applied in paleoenvironmental reconstructions. This paper combines a bibliographic review with previously unpublished data from palynofacies analyses. The aim is to present some applied examples illustrating (1) the main approaches developed for characterization of the particulate OM in surficial deposits, and (2) the study of OM transfers in terrestrial geosystems.

Keywords

organic matter; optical marker; sedimentary load; surficial deposits; soil; carbon cycle

1. Introduction

Studies of organic matter (OM) from present-day terrestrial environments (i.e. soils, surficial deposits, wetlands) are very numerous because of its sensitivity to environmental changes caused by climate and human impacts. Because OM is composed molecules containing C, H, O, N and S atoms, OM also has a major role in biogeochemical cycles through its various reservoirs and fluxes in terrestrial geosystems (e.g. Adams et al., 1990; Di-Giovanni et al., 2002).

On one hand, numerous studies are based on geochemical approaches that allow the characterization of isotopic and/or molecular markers (*biomarkers*) used for the identification of the origin of major organic compounds (e.g. Meyers, 1997; Ismaïli et al., 1999). Although these methods have been quite successful (e.g. for dissolved organic compounds), they present some limitations. For example, molecular analyses only focus on a small proportion of the whole OM (e.g. Lichtfouse et al., 1997). Moreover, the characterization of natural and anthropogenic OM by bulk chemical methods provides an averaged signal that can be difficult to exploit because of its heterogeneous composition (Sebag et al., 2006a). These technical restrictions can be sometimes circumvented (e.g. Bouloubassi et al., 1997), but they restrict the study to the OM dynamics in soil profiles (i.e. humification and mineralization processes) and wetland deposits (i.e. peat formation or alteration), and the OM transfers (i.e. fluxes of organic carbon) between the major reservoirs of the catchments.

On the other hand, many studies of the sedimentary OM from geological marine (e.g. shale, limestone) and continental formations (e.g. coal seams, shales) are based on optical analyses such as palynofacies, maceral analysis, and reflectance measurements. In fact, all these techniques are used to quantify the micrometric heterogeneity of the particulate OM (Tyson, 1995). In this way, recent works showed that the palynofacies analysis could be used to characterize the OM of present-day samples: soil layers (Di-Giovanni et al., 1999a),

lacustrine sediments (Noël et al., 2001), palustrine and alluvial deposits (Gastaldo & Huc, 1992; Cohen et al., 1999; Sebag et al., 2006b), coastal environments (Marchand et al., 2003; Sparica et al., 2005), upwelling deposits (Lallier-Vergès et al., 1993a; Lückge et al., 1996; Valdés et al., 2004; Pichevin et al., 2004) and deep-sea pelagic sediments (Lallier-Vergès & Albéric, 1990; van Waveren & Visscherb, 1994).

Thus, the palynofacies method is well known and classically applied by petroleum geologists, sedimentologists and by Quaternary scientists who study palaeoenvironments and climate changes. In order to enlarge its development, this review and further prospects is addressed to all scientists interested in particulate transfers in catchments. This paper consists of (1) a concise presentation of methodology since it is well described elsewhere (e.g. Combaz, 1964; Tyson, 1995; Batten, 1996), (2) a review of the main approaches performed for the characterization of the particulate OM from surficial deposits, and (3) a debate on possibilities to study the different ways of terrestrial OM transfers. These two latter points will successively be exploited in surficial deposits (soil profiles), elementary geosystems (lacustrine basins, wetlands) and at the catchment scale.

2. Palynofacies analysis

Palynofacies analysis is based on transmitted light microscope study of organic constituents isolated and concentrated by acid and basic digestions (Combaz, 1964). It permits quantification of various organic constituents (Tyson, 1995) and is used for the assessment of petroleum potential of source rocks, for biostratigraphical correlations, and for reconstruction of depositional environments (Batten, 1996).

The organic constituents examined under optical microscope show a large diversity in terms of morphology, color, opacity and recognizable biological structures (Tyson, 1995). Globally, the classification used in this review of a large panel of terrestrial geosystems describes several categories of particulate OM organized into three main groups (Tyson,

1995; Batten, 1996; Fig. 1). Amorphous OM (AOM; Fig 1a), which preserves some various optical, chemical and structural properties, originates from bacterial, algal or planktonic matter. The discrete elements are some recognizable organs or organisms. They are classified according to their taxonomic level and/or their morphotype: palynomorphs (SP: spores and pollen), fungal remains (MYC: mycelium fragments) and zoomorphs or zooclasts (ZOO). The Phytoclasts regroup the whole particulate constituents derived from higher plants or from their degradation (Fig 1b-f). In this group, two categories of phytoclasts are distinguished: (1) preserved fragments (CM: cuticle and membraneous fragments; TLC: transparent ligno-cellulosic fragments; ALC: altered ligno-cellulosic remains) present initial biogenic structures, and (2) transformed particles (AP: amorphous particles; GP: gelified particles; OP: opaque particles) present characteristic properties resulting from their degradation or their thermal maturation (Tyson, 1995; Batten, 1996). The plant fragments are observed in various degradation stages: transparent tissues (TLC, CM) correspond to the best-preserved state, whereas reddish altered tissues (ALC) present a more advanced degradation state, i.e. an intermediate step between TLC and AP.

This simplified classification must be considered a compromise between the various particulate organic content and the different nomenclatures encountered in the literature. In this instance, the Amorphous Particles (AP) encompass more and less amorphous constituents according to their degradation state or to the preliminary chemical treatment (Laggoun-Défarge et al., 1995). In fact, the AP constitute a continuum from particles with diffuse outlines to totally amorphous constituents, but they exhibit a typical behaviour under fluorescence. Hitherto fully unstructured constituents are finally ranged in phytoclast group insofar they are identified as deriving from terrestrial constituents (e.g. the terrestrial AOM quoted in literature). In the same way, the Opaque Particles (OP) include heterogeneous constituents that could have various origins such as combustion residues (i.e. soot and

charcoal), strongly altered and oxidized phytoclasts created by repeated cycles of degradation (i.e. charcoal-like phytoclasts) and/or a probable fraction altered by thermal maturation (i.e. coalified material) and inherited from the geological substratum (Di-Giovanni et al., 1999b and 2002; Batten, 1996; Tyson, 1995). In addition to these organic constituents, opaque mineral particles (e.g. sulphides, oxides) could resist preliminary chemical treatments. These various constituents are not easily distinguished in transmitted light mode (e.g. shape, outline aspects and possible residual structures), but can be easily discriminated in reflected light mode (i.e. maceral analysis, reflectance measurements).

3. Characterization of particulate OM

In modern terrestrial environments, two complementary approaches are classically used to characterize particulate OM from optical analyses (Tyson, 1995 and references therein). Formally, the optical markers were used for the identification of particles typical of the different sources (plankton, soils, vegetal cover) in the deposits. The optical signatures were used to characterize and compare OM content from various environments (peat, soils, sedimentary load, lacustrine deposits), with the combination of several optical markers.

3.1. Optical markers

This straightforward method is based on the use of some typical particles (*optical markers*) according to their nature (i.e. aquatic or terrestrial), their origin (i.e. biogenic, anthropogenic or fossil), and/or their formation (i.e. biodegradation, combustion, weathering). For example, this approach was performed for the correlation between changes in OM composition and environmental, climate and/or land use changes in various contexts as Alpine and Andean lakes (Buillit et al., 1997; Sifeddine et al., 1998; Noël et al., 2001), tropical and temperate wetlands (Sifeddine et al., 1995; Bourdon et al., 2000; Sebag et al., 2006b), and streams from sub-mediterranean mountains (Martaud, 2003).

3.1.1. Aquatic (planktonic) or terrestrial (higher plant) nature of OM

The optical markers are suitable for the study of lacustrine environments marked by contrasting biological inputs. In such environments, sedimentary OM comprises an aquatic fraction (i.e. direct biological inputs) and a terrestrial fraction (i.e. plant debris, reworked particles from soils). Hence, numerous works identify and estimate these two distinct stocks in recent and Holocene deposits by using optical markers and various ratios (Bertrand et al., 1992; Lallier-Vergès et al., 1993b; Di-Giovanni, 1994; Patience et al., 1995; Bourdon et al., 2000; Noël et al., 2001; Grill et al., 2002).

For example, the "Amorphous OM/Gelified Particles ratio" was used to define the differences in OM content in lacustrine and palustrine sediments and to estimate the fluctuations of water-level in a tropical maar deposit (Tritrivakely, Madagascar; 19°47'S, 46°55'E, about 1800 m a.s.l.; Sifeddine et al., 1995). These fluctuations are linked to main palaeoclimatic changes during the last 36 kyr. Aridification periods (before 36 kyr, between 25 and 13 kyr and during some episodes of the Holocene) are characterized by a peat-bog installation, while wet periods (between 36 and 28 kyr, and some episodes of the Holocene) are characterized by a lacustrine phytoplanktonic production and the occurrence of higher plants on the land.

The "AOM/Phytoclast ratio" was also used to determine the modifications of lacustrine OM induced by climate changes and human activity in temperate lacustrine deposits (Annecy, France; 45°54'N, 6°07'E, about 600 m a.s.l.; Buillit et al., 1997). This study showed that these optical markers can be used to quantify the dilution of planktonic OM (i.e. aquatic biological production) by the land-derived supplies (i.e. vegetation cover and pedogenic inputs).

More recently, the "AOM/TLC ratio" was used to characterize the modern organic fluxes and the postglacial organic sedimentation in calcareous catchment area (Chaillexon, France; 47°04'N, 6°39'E; Di-Giovanni et al., 2000a). This study shows (1) a seasonal variation in the

detrital organic fluxes directly linked to the autumnal litter production and (2) abrupt changes of OM composition in the lacustrine sedimenraty record related to the modifications of the treeline position on the watershed during the Postglacial.

In all these examples, the variations of sedimentary OM (aquatic vs terrestrial markers) are linked to fluctuations of water-level and/or terrestrial sedimentary fluxes. The applications of this kind of optical markers are frequent insofar they provide various proxies used to reconstruct paleoenvironmental changes in sedimentary basins and/or their watersheds. Furthermore, palynofacies analyses can be used to identify specific markers of the origin of terrestrial organic inputs.

3.1.2. Origin of terrestrial organic inputs

The particles considered as allochthonous and/or autochthonous may vary according depositional environments (Di Giovanni, 1994; Tyson, 1995). A study focussed on environmental and climatic changes in the French Jura mountains (Lautrey, 46°35'14"N, 5°51'50"E, about 788 a.s.l., Magny et al., 2006) found that autochthonous (i.e. lacustrine) OM can include not only phytoplankton but also particles inherited from aquatic plants, while allochthonous (i.e. terrestrial) OM encompasses particles weathered from catchments and/or windblown grains. This study shows that terrestrial OM is predominant in Postglacial lacustrine deposits. Hence, the OM sedimentation principally reflects the development of soils and vegetation on the lacustrine basin and the catchment area.

Some phytoclasts serve as optical markers provide that a potential tool to track (1) biological terrestrial inputs (TLC) from local plant cover or reworked from fresh forest litter; (2) the pedogenic fraction (AP) reworked from the surrounding soils and (3) various allochthonous mature inputs (OP) including carbonized (i.e. charcoal, pyrofusinite) and geological (i.e. fossil OM contained in sedimentary rocks) particles (Lallier-Vergès et al., 1993b). In addition, gelified particles (GP) are often observed in hydromorph soils and

correspond to an early gelification of the cell luminae products of root tissues. These gelified tissues are further divided into gelified debris with a homogeneous texture and with a polygonal morphology (Lallier-Vergès et al., 1998; Noël et al., 2001). The relative contributions of these complementary markers have been used to characterize the relations between terrestrial organic fluxes and climatic variations in various environments.

In a temperate lacustrine basin (Le Bouchet, France; 46°57'N, 0°09'E, about 1200m a.s.l.), terrestrial optical markers have been used to identify major changes of organic terrestrial fluxes occurring during the last 15 kyr (Lallier-Vergès et al., 1993b; Sifeddine et al., 1996). The variation of these fluxes showed: low organic fluxes during the Lateglacial, an increase at the beginning of the Holocene, a minimum at the end of the Atlantic period resulting from the climatic cooling, and a maximum at the end of the Sub-Boreal related to the installation of the present climatic conditions.

In other Alpine lacustrine basin (Lac d'Annecy, France ; 45°54'N, 6°07'E, about 600 m a.s.l.), palynofacies have been used to. the palynofacies composition have been used to identify different terrestrial organic sources including forest floors, soil-horizons and geological substratum (Buillit et al., 1997; David et al., 2000 ; Noël et al., 2001). These studies showed that the relative variations in organic sources are directly dependent on human land-use. For example, Noël et al. (2001) showed extreme events such as flood or intensive run-off are characterised by notable increases of organic matter from surface berween 5000 and 1700 BP (i.e. low human impact) and deep soil layers since 1700 BP (high human impact).

In residual lakes in a "cloud forest" (Central Andes, Bolivia; 17°50'S, 64°43'W, about 3000m a.s.l.), terrestrial optical markers (CM, TLC, AP, GP, OP) have been used to identify major environmental changes during the last 15 kyr: high detrital fluxes during the Last Glacial Maximum, development of vetation cover and soils during the Postglacial and

increasing charcoal contents related to aridification and paleofires between 8.8 and 4.5 kyr BP (Sifeddine et al., 1998). In rainforest (Eastern Amazonian, Brazil; 6°35'S, 49°30'W, about 600m a.s.l.), the results of geochemical and palynofacies analyses lead to a reconstruction of the variations in the regional hydrological regime over the last 30 kyr. (Sifeddine et al., 2001).

In tropical humid context (Lagoa do Caço, Brazil; 2°58'S, 43°25'W, about 120m a.s.l.), optical analyses are exploited through the debate on source and degradation conditions of terrestrial origin particles (Jacob et al., 2004). Bulk organic geochemistry and petrography are combined with sedimentological evidence in order to identify the major environmental changes that occurred since 20 kyr BP. In this study, the results emphasise the main advantage of palynofacies allowing identification of specific or scarce organic constituents for which conventional geochemical data are sometimes unsuitable.

3.1.3. Identification of fossil organic contributions

The optical marker approach has also been used to elucidate the presence of fossil OM in soils, lacustrine deposits, fluvial and marine sediments. The fossil OM observed in all compartments would be inherited from the sedimentary rocks outcropping in the catchment. Some coalified particles reworked from ancient formations have been observed in recent deltaic sediments (Rhone, France; 43°18'N, 5°24'E; Gadel & Ragot, 1973). In catchments presented in Figure 2, fossil OM has even been recognised in the whole of soil profiles. In addition the relative contribution of these fossil particles is closely related to the amount of sedimentary OM in the geological substratum (Fig. 2b).

In the Chaillexon lake catchment, some angular OP were observed in the present-day humus layers (Fig. 2c1; Di-Giovanni et al., 1998), in the lacustrine deposits and even in calcareous Meso-Cenozoic formations (Di-Giovanni et al., 1997). These works emphasise the significant contribution of fossil OM in terristrial organic fluxes and its disappearance when

soil and vegetation cover became efficient enough to protect the bedrock from mechanical erosion (i.e. at beginning of the Boreal period about 9000 yr BP).

In the marly catchment of Les Peyssiers (France; 44°34'N, 6°05'E, about 850 m a.s.l.), OP reworked from Mesozoic substratum constitutes the majority of the particulate OM in the organo-mineral soil layers (50 to 70%; Fig. 2c2) and in the present sediments (Di-Giovanni et al., 1999b). These results show that OP must not be systematically interpreted as degradation product of present-day or recent biologic production (i.e. oxidation or combustion). Moreover, the total amount of OM in soils and sedimentary records not only depends in the net primary production but also on erosion processes of bedrock. Thus, this work suggests that past alluvial and lacustrine OM not only gives evidence of past vegetation cover but can reflects detrital inputs from the geological sustratum.

In the marly catchment of Draix (France; 44°13’N, 6°35’E, between 850 and 1250 m a.s.l.), OP are also identified in organo-mineral soil layer and are dominant in sedimentary load from the streams (Martaud, 2003). Here again, OP are considered as fossil markers and besides, this study highlights that the fossil contribution in soils and rivers is controlled by the total organic carbon (TOC) of the underlying bedrock and by the density of vegetal cover. These parameters act as positive and negative feedback respectively to the contribution of fossil organic matter in soils.

In all these study cases, OP are identified as being the major fossil contribution. However, the fossil OM markers are related to the characteristics of the parent-rocks. In fact, it is necessary to characterize precisely the organic contents from the distinct compartments of the watershed (i.e. primary reservoirs: vegetation cover, soils and geological formations) before any study of sedimentary fluxes (i.e. transfers of particulate matter: suspended load and bedload) and sedimentary records (i.e. secondary reservoirs: alluvial and lacustrine deposits).

3.2. Optical signature and palynofacies composition

This approach is based on the use of binary or ternary diagrams are often used to define the typical signatures of optical assemblages from the relative proportions of significant organic constituents. Tyson (1995, p. 441) points out that the main advantage of ternary diagrams is that they provide a spatial separation that is useful for grouping samples into empirically defined associations or assemblages. Hence, this approach is very suitable for the comparison of numerous samples from various geosystems when the same organic particles are present in different proportions in all compartments (sources, recipients). As an example in the Faroe Islands (Roncaglia, 2004), changes in relative abundance of sedimentary OM are related to changes in hydrographic parameters. In this study, the author uses the relative proportions (1) of phytoclasts and sporomorphs to determine the influence of terrestrial inputs, (2) of AOM to determine oxic/anoxic regime and (3) of zoomorphs to reconstruct the distance from the shoreline. Furthermore, it is successfully applied to track the changes in the OM composition during the humification in soil profiles (Di-Giovanni et al., 1998), the diagenesis of peaty deposits (Bourdon et al., 2000), the transport of reworked terrestrial particles (Di-Giovanni et al., 2000b; Sebag et al., 2006b) or the weathering of geological substratum (Di-Giovanni et al., 1997; Martaud, 2003).

This kind of studies based on optical markers and petrographical signatures improves the knowledge of present-day and modern environments. These progresses lead to the establishment of the relationship between the OM composition (*palynofacies*) and the depositional environments and are naturally applied for paleoenvironmental reconstructions (e.g. Tyson, 1995; Gastaldo et al., 1996; Hofmann & Zetter, 2005).

3.2.1. Characterization of soil OM in temperate environments

Optical analyses of soil OM remain scarce, generally restricted to local studies without any connections with the others, and serve to track the variation of terrigenous sedimentary

loads through the recognizance of specific markers. However, such observations would be extended and compiled, insofar these works are based on soils sampled in various climatic and geomorphological settings (Di-Giovanni et al., 1998; Di-Giovanni et al., 1999a and b; Noël et al., 2001; Sebag et al., 2006b).

Figure 3 shows that the main compartments of soil profiles can be identified by the comparison of the relative contributions of preserved phytoclasts (CM, TLC, ALC) and transformed particles (AP, GP, OP; Fig. 3a). For plant litter, a high amount of preserved phytoclasts (between 60 to 75%) accompanied by 15 to 35% of transformed particles are typical of "biological" signatures from OL horizons. The OF and OH horizons present usually a "pedogenic" signature marked by 15 to 45% of preserved phytoclasts and by 40 to 70% of transformed particles. For organo-mineral layers (i.e. A horizons) the contents of transformed particles are obviously anti-correlated to those of preserved phytoclasts: low (about 50%) in the surficial layers supplied by fresh plant tissues (e.g. roots, cuticles), dominant (up to 99%) in the deeper layers enriched in fossil OM (i.e. OP in the samples studied). Finally, in the organo-mineral horizons, the OM composition corresponds to a blending curve between two contrasting poles corresponding to surficial and deep layers (Fig. 3a). For forest soils, surficial layers exhibit litter-like signatures while deeper layers more a humic-like ones. In contrast, for the grassland soils, the deepest layers are characterized by typical signature with more than 90% of transformed particles.

Complementary, the ternary diagram "TLC/AP/OP" (Fig. 3b) reveals the characteristics of soil layers. As previously seen, the plant fragments (TLC) are characteristic of the OL horizons and represent direct biological inputs. On the other hand, the "AP contents" are dominant in the other organic layers (OF, OH). Previous studies of soil OM (Di Giovanni, 1994; Maman, 1997) have shown that pedogenic degradation might lead to the amorphisation of ligno-cellulosic fragments. Similarly, the degradation of higher plant debris from

Cyperaceae leads to the formation of amorphised particles (AP) in a tropical peatland (Bourdon et al., 1997) and in a modern mangrove swamp (Lallier-Vergès et al., 1998). According to Noël et al. (2001), the proportions of the preserved (TLC) and altered (ALC) phytoclasts and the amorphised particles (AP) in lacustrine deposits may be considered as source-markers: forest litter (TLC), surficial organic soil layers (ALC), or deep organic soil layers (AP). As shown previously, the fossil OM (i.e. OP in Figure 3) is more abundant in the organo-mineral (A horizons) and geological layers (alterite C; Fig. 2 and 3b).

In detail, some OF and OH samples exhibit a "plant litter" signature (Fig. 3a) and some OF and OH samples and some grassland and forest A samples exhibit a similar signature (Fig. 3b). These points reveal the OM characteristics depend on the observation scale. On a larger scale, the classification of organic horizons bears on the macroscopic observations (physical characteristics) related to the transformation of organic constituents. On a smaller scale, these transformations are linked with some molecular and/or elementary analysis (chemical characteristics). On an intermediate scale, microscopic observations (optical characteristics), related to particulate OM, could provide complementary information between macroscopic and geochemical investigations.

Applied to the soil samples, the palynofacies analysis allows distinction between the main soil horizons (Di-Giovanni, 1994; Buillit et al., 1997; Noël et al., 2001). Comparison of means and confidential intervals (Fig. 4a) shows the differences between the main soil layers. A simple statistical analysis (ANOVA; Fig. 4b) shows that the more significant differences are related to the occurrence of preserved phytoclasts (TLC) and transformed particles (AP and OP). Indeed, their formation and preservation depend on the local context (i.e. hydromorphy, geological substratum, periodic tilling) and the local plant cover (i.e. nature of fresh inputs, biological productivity).

These analyses of soil OM are restricted to the validation of some previous results and not enhance our knowledge. Nevertheless, they can be considered as a pertinent tool for the characterization of continental organic reservoirs and above all, find its interest for the survey of the evolution of OM stocks when classical investigations, as bulk geochemistry, are inefficient.

3.2.2. Depositional environments of particulate OM in alluvial wetlands

The palynofacies analyses also provide information about the distribution of OM and sedimentary dynamics in present-day terrestrial geosystems (e.g. Gastaldo & Huc, 1992; Tyson, 1995; Gastaldo et al., 1996). For example, an inventory of particulate OM in a modern alluvial wetland (Vernier Marsh, France; 49°25'N, 0°27'E, about 4 m a.s.l.) showed the spatial distribution of the main organic (Sebag et al., 2006b). Thus, the composition of the sedimentary OM is specified for each studied compartment (i.e. soils, drainage ditches, channels, ponds) using two ternary diagrams (Fig. 5). The "AOM contents" increase from terrestrial environments (i.e. soils and histosols) to fluvial deposits (Fig. 5a) and reflect the abundance of aquatic production. The "Preserved/transformed phytoclast ratio" decreases from production (i.e. litters) to deposition area (i.e. ditch, pond and alluvial deposits; Fig. 5a) indicating the various degradation stages of terrestrial inputs. "OP contents" allow definition of fluvial (high values) and palustrine (low values) signatures, while "GP/AP ratio" allows to identification of soil (low values) and plant supplied samples (high values for pond deposits; Fig. 5b).

On the other hand, the study of modern and Holocene deposits allowed identification of the relations between palynofacies and palustrine depositional profiles in riparian backswamps (Fig. 6; Sebag, 2002). All the particulate constituents are present in fluvio-palustrine deposits, but their relative abundances are different according to the depositional environment. In the aquatic environments (i.e. river, pond, pool), particulate OM groups into a

major autochthonous fraction, i.e. algal or planktonic amorphous constituents (AOM), and a minor allochthonous terrestrial fraction, i.e. more or less preserved plant fragments (phytoclasts). In the peaty wetlands (i.e. fen, wet meadow, swamp), the autochthonous plant-derived constituents represent the main organic fraction, but some amorphous constituents can reflect a high groundwater-level (i.e. autochthonous AOM in closed peatland) or significant fluvial inputs (i.e. allochthonous AOM and OP in alluvial marsh). Moreover, the preserved debris (TLC) and gelified fragments (GP) are abundant in aquatic conditions (e.g. fen, wet meadow), even though the altered fragments (ALC) and amorphised particles (AP) are dominant in terrestrial conditions (i.e. histosols). In the forest soils, pedogenic fraction (ALC, AP) is naturally dominant, but the preserved fragments (TLC) can be abundant in relation to a high productivity of the local plant cover. In addition, the abundance of the fungal fragments (MYC) is a usable marker to the aerobic biodegradation of plant remains.

3.2.3. Distribution of particulate OM in catchments

Palynofacies analyses also provide the optical composition of particulate OM of the main compartments of catchments (Di-Giovanni et al., 2000b). This study is based on the characterisation of suspended load and bedload OM and the comparison with the palynofacies of the geological formations outcropping on the Moulin River's watershed (Draix, France; 44°13'N, 6°35'E). The results emphasise the contribution of reworked fossil OM in the modern detrital fluxes.

The Lower Seine Valley is also a good example. This catchment includes various geomorphological compartments (Fig. 7a; Laignel et al., 2002; Sebag, 2002; Laignel, 2003): a chalk substratum (i.e. Upper Cretaceous), a Cenozoic weathering cover (i.e. clays-with-flints and slope clays-with-flints) and a Pleistocene sedimentary cover (i.e. loess deposits on plateaus). These ancient geological elements are overlayed by some soils and alluvial (i.e. coarse fluviatile deposits) and palustrine (i.e. peaty or clayey deposits) valley formations aged

to Late Quaternary and Holocene periods (Fig. 7a). As seen in sections 3.2.1 and 3.2.2, the OM compositions from the various organic samples, i.e. higher plant supplied compartments as soils and palustrine deposits, are marked by differences related to the changes of the OM in the forest litter, the humic soil layers, and in the peaty deposits (Fig. 6 and 7). However, the palynofacies from each type of geological sample (i.e. chalk and clays-with-flints) are very uniform and dominated by a specific amorphous constituent ("sub-colloidal" OM in Combaz, 1964). Palynofacies of marls and limestones from Draix catchment contain an identical amorphous constituent that is revealed to be a mixture of clayey minerals and undetermined organic compounds (Martaud, 2003). Except for this unidentified amorphous constituent, Opaque Particles (OP) dominate the palynofacies of the substratum from the Lower Seine catchment, although there are some minor differences (i.e. TLC, AP, GP contents). This “OP-palynofacies” is similar to other geological substrata (Di-Giovanni, 2000c; Di-Giovanni et al., 1998; Noël et al., 2001) and can be proposed as typical of marls and limestones. Furthermore this palynofacies is also similar to that of deep soil and alterite layers (see sections 3.2.1 and 3.2.2) and sedimentary load from streams (Martaud, 2003).

Palynofacies method finds its limitation when it is suspected that the studied reservoirs, frequently a soil, is rich in combustion residues (Schmidt et al., 1999) and for which particular fossil OM and black carbon (BC, pyrofusinite) should be separately distinguished. In such case, but, to our knowledge, it was never tested , a complementary method in reflected light mode, as reflectance and maceral analyses, classically used by coal petrographers, would be performed in order to avoid some confusions and surestimates of BC stock in soils or in marine sediments as suggest Dickens et al. (2004).

4. Transfers of OM in continental geosystems

4.1. Transformation of OM in the soil profiles

Previous studies of the particulate OM in soil profiles focused on the transformations of fresh biological inputs and the transfers of fossil OM. The palynofacies method will not provide new fundamental information on OM transfers from biosphere to pedosphere (i.e. mineralisation) and to the related chemical changes (i.e. humification), but it focuses on the particulate by-products of pedogenesis. In addition, this method considers the particulate soil OM as a continuum from initial biological inputs to ultimate pedogenic residues. Moreover, optical analyses provide some relevant arguments on OM transfers from the geosphere to the pedosphere (see section 3 for further details).

As previously shown (Fig. 2iii), this approach has been used to identify the main changes in soil OM composition and to follow (1) the progressive decrease of biological constituents (LCT) from surficial to deep soil layers related to degradation processes; (2) the resulting increase of pedogenic constituents (AP) in the humic layers; and (3) the relative increase of fossil OM (i.e. OP in samples studied). This fossil contribution results in a direct incorporation into organo-mineral and mineral layers related to the chemical weathering of geological substratum. Following the example of Draix catchment (Copard et al., 2006), fossil OM incorporation into soils appears to be mainly driven by two parameters (1) the initial organic carbon content of the underlying bedrock (marls or marly limestone; Fig. 2b), and (2) the density of vegetation cover (Fig. 8). If the former merely depends on the geological setting, the latter is related to climatic conditions, which may or may not promote consequent biomass production and microbial consumption. In addition, the palynofacies analyses have been used to study the indirect contribution (i.e. lateral supplies) resulting to a mechanical weathering and related to topographical controls (Di-Giovanni, 2000c).

4.2. Organic fluxes in wetlands, lakes and rivers

When applied to modern alluvial wetlands, this approach helps to understand and to track OM transfers at the geosystem scale (Sebag et al., 2006b). For instance, the optical signatures express an AOM negative gradient from terrestrial stations (histosols) and soil-derived samples (ditches) to pond and river deposits (Fig. 3a) as a result of an increasing aquatic contribution. In flowing water environments (i.e. ditch, channel and river), the proportions of opaque particles (OP) gradually increase from drainage ditches to fluvial deposits (Fig. 3b). This evolution is accompanied by a relative increase of the aquatic component (AOM) and can be related to an increasing alteration of the terrestrial contribution during its transport (Tyson, 1995). Such information is essential to correctly decipher bulk geochemical data, which average results from mixed constituents.

Previous studies of lacustrine deposits focused on organic sedimentation as a response to environmental changes or human impact (Lallier-Vergès et al., 1993b; Di-Giovanni 1994; Buillit et al., 1997; Di-Giovanni et al., 2000a; Noël et al., 2001). They have demonstrated the millennial variability of lacustrine organic sedimentation controlled by the productivity of plant cover in the catchments. During the Holocene, in temperate regions, warm periods mostly record a greater contribution of well-preserved higher plant debris (i.e. development of forests on the catchment) and soil supplies, while cold periods instead show a greater algal contribution relative to higher plant debris (i.e. arid conditions on the catchment). The latter conditions lead to the sedimentation of very oxidized algal OM (opaque AOM). In some cases, pluri-secular climatic variations of smaller amplitude may also be recorded, as "Little Ice Age" or industrial-time warming (Buillit et al., 1997; Noël et al., 2001). Furthermore, study of modern suspended loads sampled during one seasonal cycle reveal an annual variability of particulate organic fluxes related to the autumnal forest litter supply (Di-Giovanni et al., 2000a). All these studies suggest a possible decoupling between organic

sedimentary records and climatic fluctuations. This decoupling highlights the occurrence of an environmental filter (e.g. timberline position, local plant cover, agrarian practices) as threshold, buffer and oasis effects impacting the detrital organic fluxes. As regards the threshold effect, particular attention must be paid to the high sensitivity of detrital organic fluxes to increasing human activities. For example, land changes (deforestation, cultivation, soil establishment) directly influence soil cohesion and its ability to be reworked and hence induce quantitative and qualitative fluctuations of the organic fluxes during erosion of the catchment (Noël et al., 2001; Di-Giovanni et al., 2000a).

Riverine samples (i.e. suspended and bed loads) from Draix catchments exhibit a palynofacies composition similar to those of the substratum (marls and limestones), supporting the notion that particulate OM collected at the outlet of these drainage basins essentially stems from sedimentary rocks without any qualitative and quantitative changes in fossil OM (Martaud, 2003). Furthermore, Copard et al. (2006) have shown that geochemical data for riverine particles were similar to those of marls. In this instance, gully erosion of marls ruled transfers from continental surface to streams and is considered to be the main process reloading fossil OM in streams (Di-Giovanni et al., 1999b). Therefore, these optical characterisations provide some strong arguments either about the input of fossil OM in river, or about the major impact of the erosion on the OM products transferred into fluvial networks.

4.3.Transfers of particulate OM in catchments

A large part of previous works (Patience et al., 1995; Di-Giovanni, 2000c; Noël et al., 2001) have emphasized surficial transfers related to primary productivity (i.e. aquatic or terrestrial biological inputs) and erosion of continental surfaces (i.e. mechanical weathering of soils and geological substrata). Other works have debated the involvement of chemical weathering of sedimentary rocks on fossil OM delivery in soil profiles and river networks (Di-Giovanni et al., 1999b; Martaud, 2003).

Both the definition of optical markers and characteristic palynofacies from the various compartments can be considered as some pertinent alternatives to identify and quantify the current organic fluxes in catchment areas. For example, Di Giovanni et al. (2000b) showed that studied bedload OM mainly originates from erosion of marls outcropping in the Moulin River's catchment, while the studied suspended load OM derives from erosion of recent colluvial formations. These complementary approaches have been coupled for better knowledge of the patterns of main organic transfers in mountainous context (Draix, SE France; Fig. 8; Martaud, 2003), deltaic environments (Mahakam River, Indonesia, Gastaldo & Huc, 1992; Rajang River, East Malaysia, Gastaldo et al., 1996) and estuarine catchment (Lower Seine Valley, NW France; Fig. 9; Sebag, 2002; Laignel, 2003).

In the Draix catchment, distinct reservoirs have been characterised according to the predominance (1) of preserved phytoclasts (CM, TLC, ALC) in organic reservoirs (surficial soil layers), (2) of transformed particles (AP, GP, OP) in organo-mineral reservoirs (deeper soil layers) and (3) of fossil OM (OP) in geological reservoirs and by-pass compartment (sedimentary loads; Fig. 8). In soil profiles, we discriminate (1) a primary reservoir consisting of organic horizons (OL, OF, OH) in which pedogenic processes promoting a decrease in TLC and a concomitant increase in AP and (2) a secondary reservoir consisting of organo-mineral horizons (A) supplied by plant-derived particles produced during pedogenesis and by fossil OM inherited from the weathering of underlaying sedimentary bedrock (see sections 3.2.1 and 4.1). In addition, by-pass compartments exhibit an "OP-palynofacies" pointing to a dominant OM-origin from geological reservoirs (Fig. 8). However, some slight differences between by-pass compartments and geological reservoirs still reflect soil supplies (AP, GP) in sedimentary loads.

In the Lower Seine Valley, the wide variability of OM (Fig. 9a) contents in organic surficial deposits (i.e. peat, soil litter) is attributed to local plant cover and other pedogenic

controls (e.g. hydrology, topography, climate, land uses). In contrast, the geological reservoirs (i.e. chalks, clay-with-flints) present a homogenous and typical "OP-palynofacies" (except "sub-colloïdale OM", see section 3.2.3). Some small differences in terms of contributions and size of various constituents may occur according to the impact of chemical weathering (i.e. clay-with-flints, organo-mineral soil layers) or the solifluction process (i.e. slope clay-with-flints) affecting these geological reservoirs (Fig. 9b and c; Laignel, 2003; Sebag et al., 2003). Finally, both organic (i.e. peat, soil litter) and geological reservoirs release some different detrital fluxes that may mix with autochthonous OM from aquatic primary production in fluvial and alluvial deposits (Fig. 9b).

However, the relative contribution of both these reservoirs delivering allochthonous OM depends on various factors. Thoroughly and according to the previous results, we can argue that input of fossil OM in river networks and its transfers through a specific catchment is rather significant provided that the catchment (1) is underlain by OM-rich sedimentary rocks, (2) has a scattered plant cover, (3) is subject to mechanical weathering encouraged by the combination of active tectonics and rainfall (Di-Giovanni et al., 1999; Copard et al., 2006), (4) presents an ideal geomorphology (e.g. slope, erodibility) favouring a fast and large transfer of sedimentary loads and (5) exhibits a fast turnover of its components in soils and rivers. Finally, fossil OM contribution can be significant and even major in present-day particulate organic transfers (Martaud, 2003). It may have a role which was, until now, been underestimated in the global carbon dynamics (Aitkenhead & Mc Dowell, 2000). As suggest Copard et al. (2006), this latter factor points out the importance of residence time governing OM turnover in the different reservoirs: for example, in soils, time acts as a positive feedback on the mineralisation extent of fossil OM caused by chemical or biochemical media. In addition to these extrinsic factors, it is essential to encompass those that are intrinsic. They are related to the chemical composition and physical structure of OM and act a major role in the

ability of OM to be mineralised before its deposition in a sedimentary area. Both chemical and physical features of fossil OM depend on the maturity level that was reached and consequently vary according to the geological setting. Conversely, input of OM inherited from soils and peatlands in river networks will be dominant in allochthonous fluxes if the catchment (1) is formed by eruptive formations or organic-poor sedimentary rocks, (2) has a dense plant cover or wetlands, (3) is subject to chemical weathering favored by specific climatic conditions leading to soils development and (4) presents a low turnover. In addition, transfers of allochtonous OM in and through the river networks depends on the whole set of these factors, which themselves depend on the climatic and/or tectonic patterns. Their good knowledge is the key component if we want to understand how the transfer is achieved and why the contribution of allochthonous OM can be significant in sedimentary loads and further, in surficial sediment of any depositional systems (lakes, estuaries, continental shelves).

5. Conclusions

This bibliographic review and some previously unpublished data show the real potential to exploit optical analyses in order to study organic transfers in present-day terrestrial environments. This technique provides various tools, which were successfully used to characterize the organic fluxes in surficial deposits, soil profiles, wetland and lacustrine ecosystems, and within catchments. Coupled with bulk geochemical analyses and radiocarbon data, it allows revaluation of OM origins, reservoirs, and fluxes in soils, sedimentary loads carried out by river networks and recent deposits.

Beyond these basic applications, the palynofacies method provides a valuable contribution connecting particulate OM composition to depositional environments. In addition, the palynofacies method can be used to quantify terrestrial organic fluxes and to provide some guidelines to understand how particulate transfers occur in soil profiles and

through catchments. This fundamental information is essential for a realistic modelling of terrestrial organic carbon fluxes from digitalized elevation maps. Finally, these modelling would combine the nature of plant cover and geological mapping, as well as topographical information.

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Figure captions

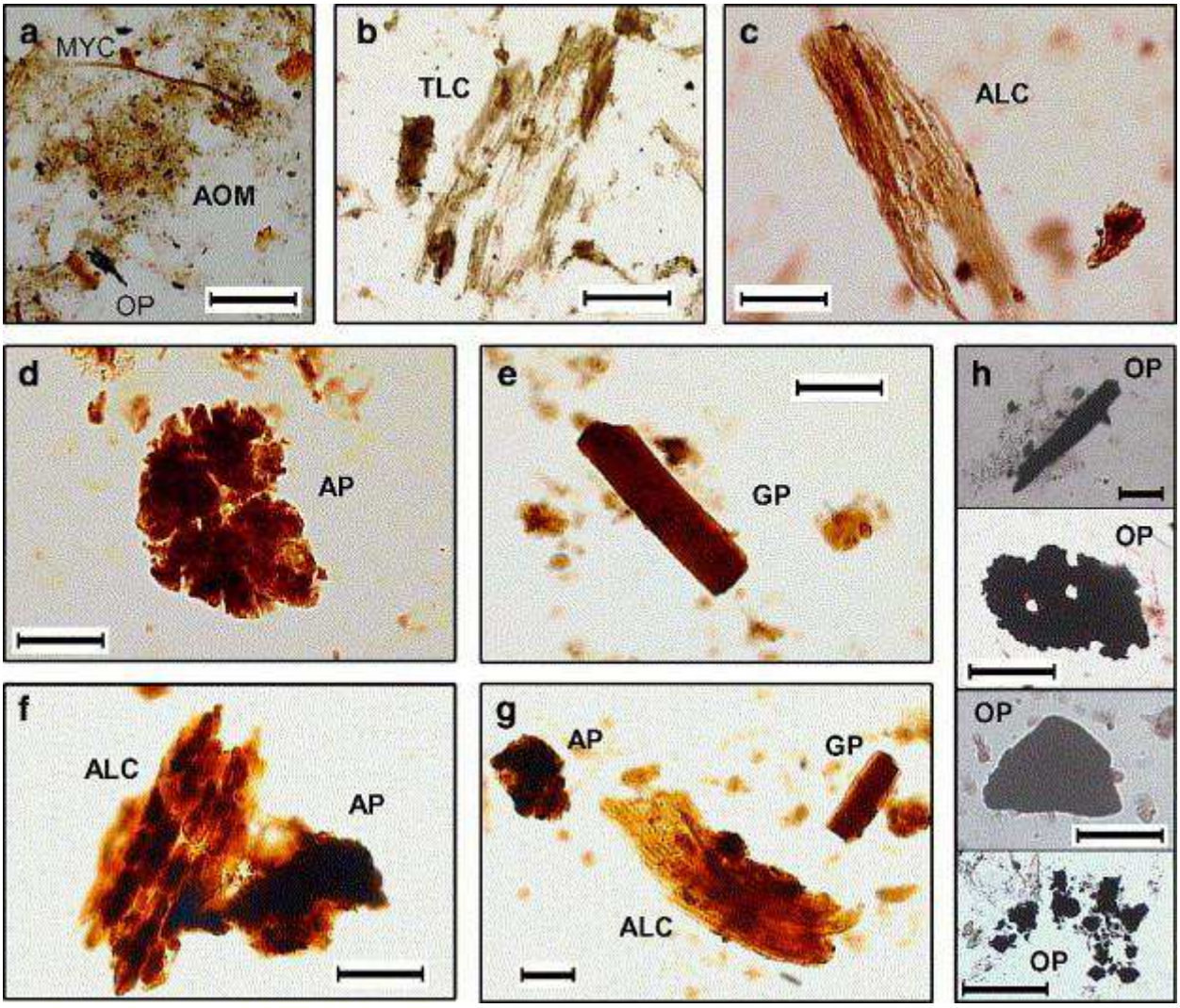


Figure 1: The main categories of particulate OM distinguished by their morphological criteria: (a) Amorphous OM (AOM) presents a flaky shape, a lumpy or granular texture, diffuse outlines and variable size, opacity and colour. (b to g) Various phytoclasts resulting from degradation of the plant fragments. (b) The transparent fragment (TLC) presents well-preserved biogenic internal structures. (c) Reddish altered fragments (ALC) present dulled outlines, reddish colour and more or less altered internal structures. (d) Reddish amorphised particles (AP) appears as reddish flakes with diffuse but recognizable outlines and, sometimes, residual internal structures. (e) Gelified particles (GP) present homogeneous texture, variable colour (brown to amber), true outlines, and usually angular shape but,

sometimes, dulled angles. (f and g) Various particles resulting from degradation of the plant fragments. (h) Opaque particles (OP) include various constituents according to the general shape, outlines aspects and possible residual botanical structures.

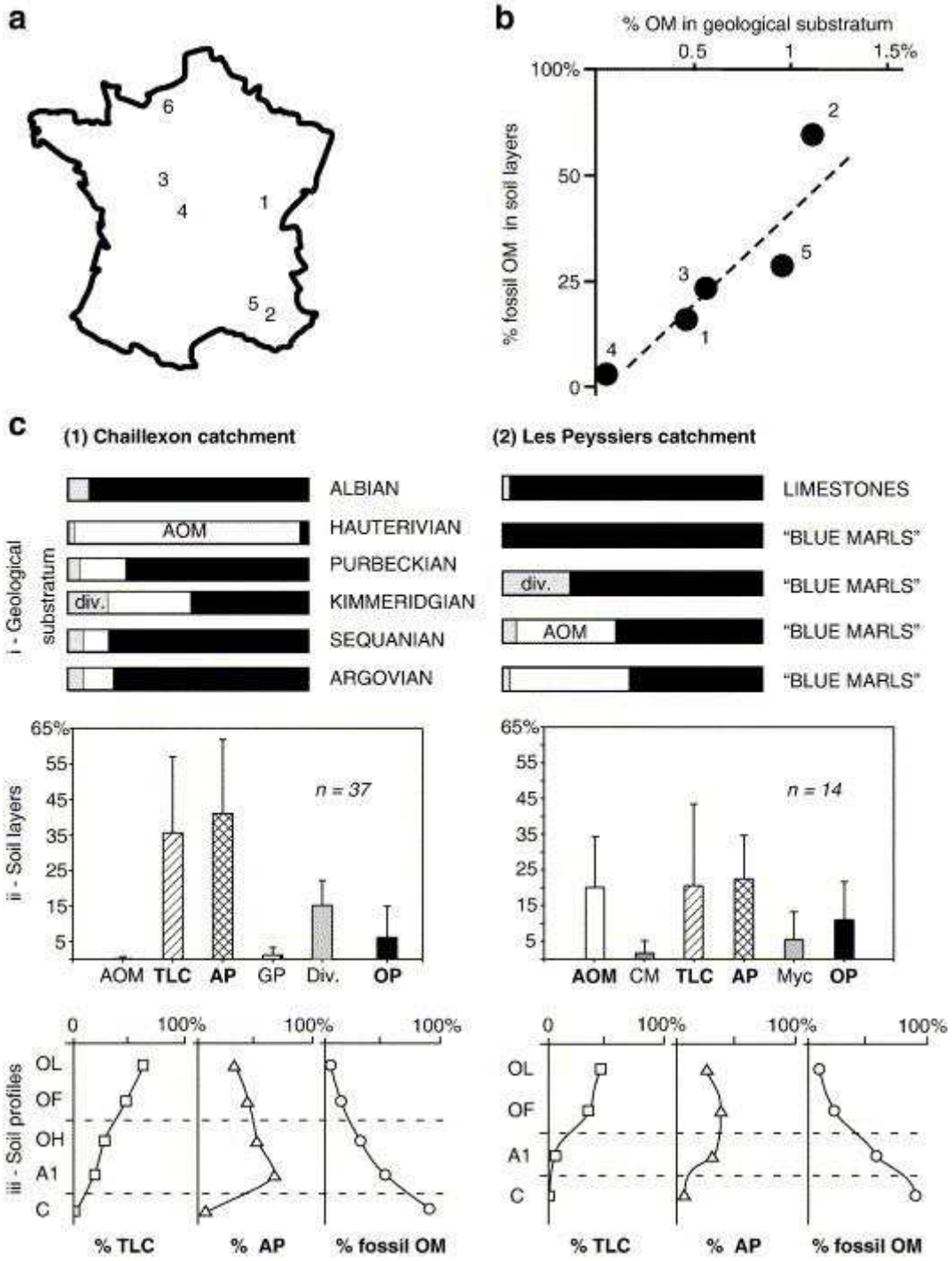
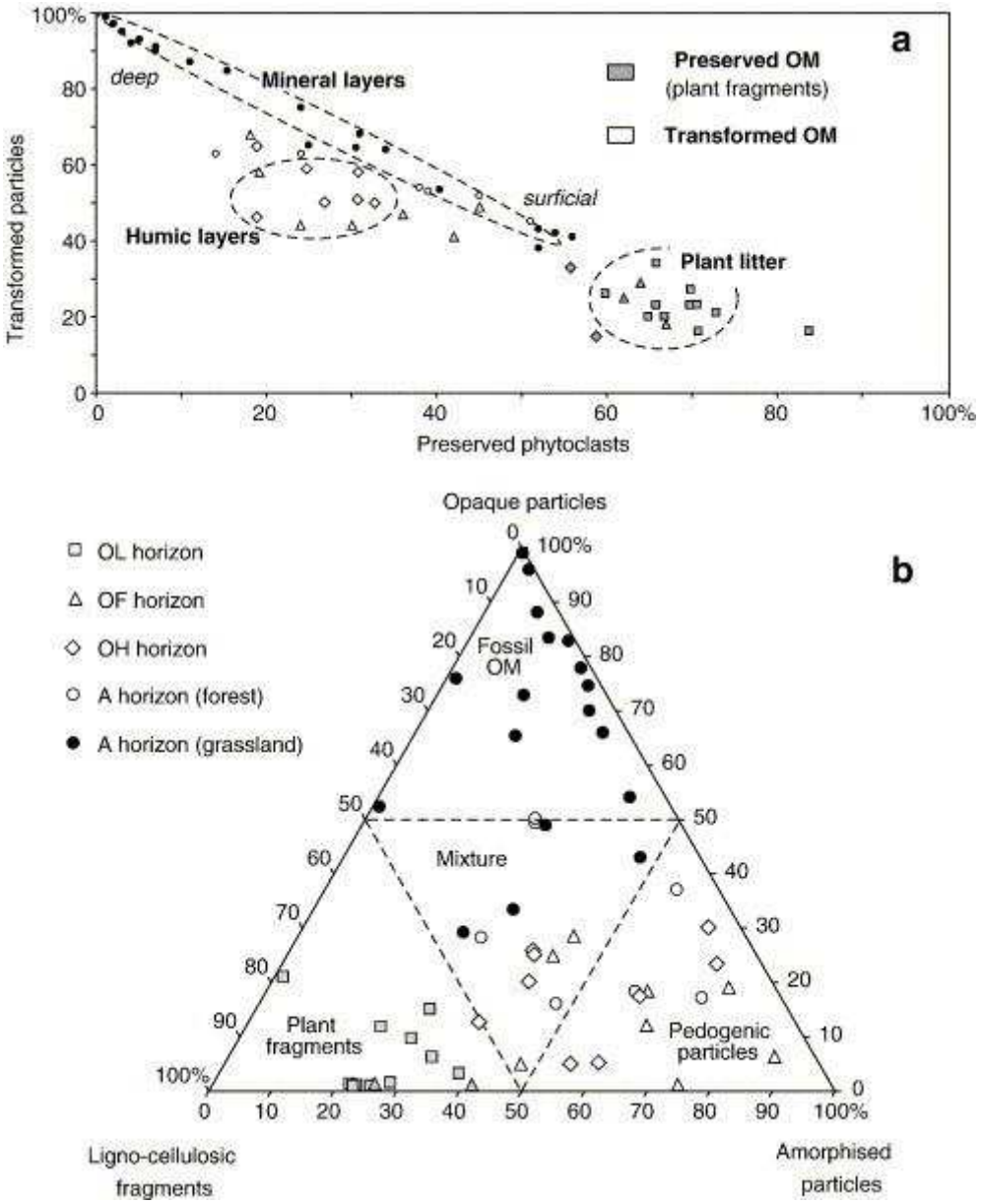


Figure 2: Study of soil OM dynamics by optical markers (from Di-Giovanni, 2000c). (a) Location of the sites: 1. Chaillexon Lake, Jura; 2. Draix catchments, Alpine moutains;

3. Négron Valley, SW Paris Basin; 4. Indre Valley, S Paris Basin; 5. Claps catchment, Prealpine mountains; 6. Lower Seine Valley, NW Paris Basin. (b) Relationship between the OM contents in the geological substratum and the fossil OM contribution in soil layers from five temperate catchments. (c) Comparaison between the (1) Chaillexon and (2) Draix catchments. (i) Characteristic palynofacies of the main sedimentary formations from geological substrata. (ii) Relative contribution (mean and standard error) of the main organic particle classes in the soil layers. (iii) Evolution of three major optical markers in the soil profiles: biological input (TLC), pedogenic fraction (AP), and fossil contribution (OP).

Figure 3: Characterization of soil OM by petrographical signatures in five temperate catchments (see Fig. 2). (a) Relationship between preserved phytoclasts and transformed particles in the plant litter, the humic layers, and the organo-mineral layers. (b) "TLC/AP/OP" ternary diagram used to discriminate the nature and the origin of the main organic fractions in various soil layers.



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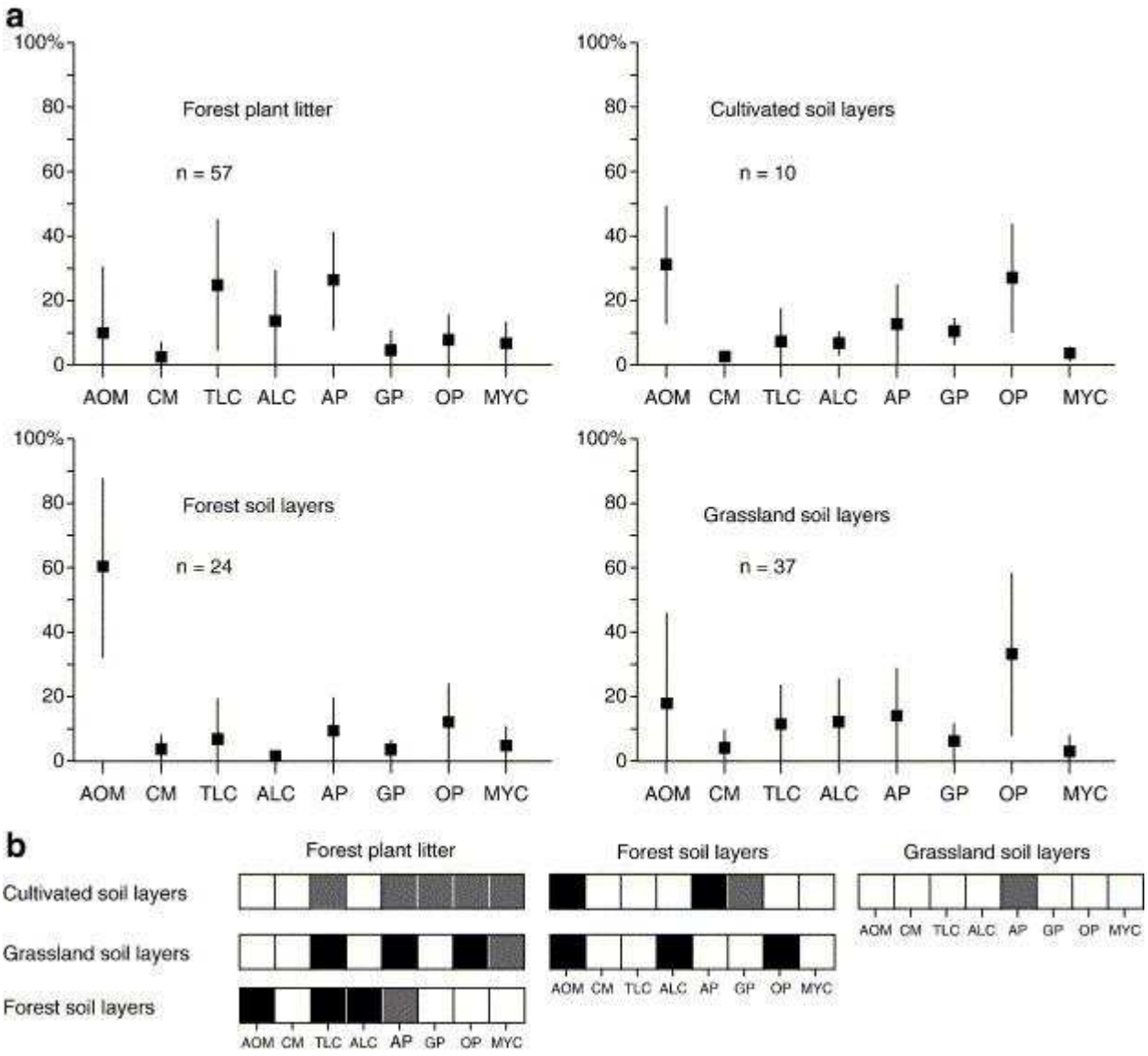

Figure 4: Composition of OM in forest plant litter and organo-mineral soil layers, in cultivated soil layers, and in grassland soil layers. (a) Mean and confidential interval from various bibliographic data (see in the text). (b) Results of statistical analysis (ANOVA): comparison for distinct soil layers. Black: significant difference with the four complementary tests (Tuckey, Fisher, Bonferroni, Dunn-Sidak). Gray: significant difference with at least one complementary tests.

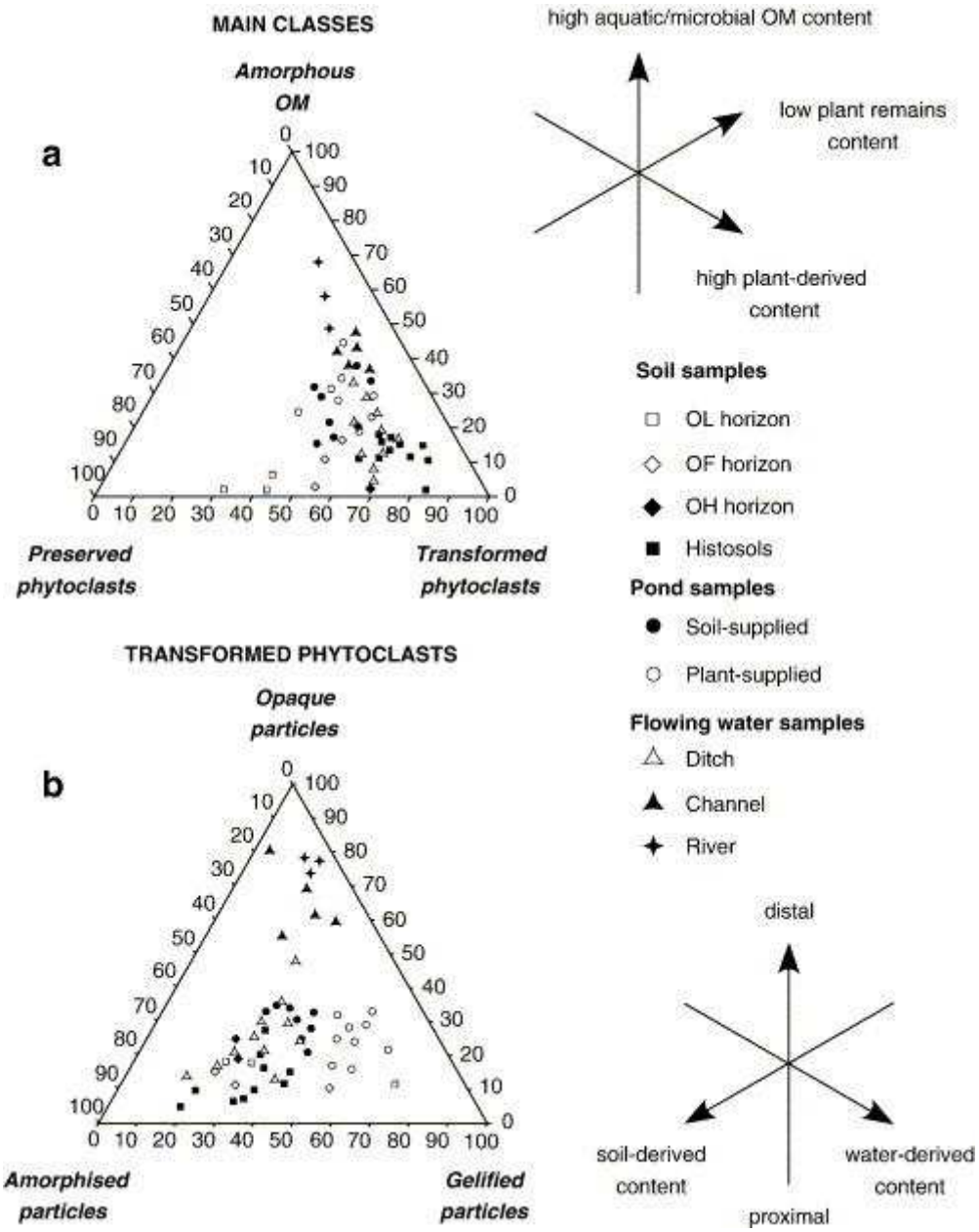

Figure 5: Characterization of sedimentary OM in wetland environments (Vernier Marsh) by petrographical signatures. (a) "AOM/Preserved phytoclasts/Transformed particles" and (b) "AP/GP/OP" ternary diagrams used to discriminate the nature and the origin of the main organic fractions in soils samples, in pond deposits, and in ditch, channel, and alluvial deposits (modified from Sebag et al., 2006b).

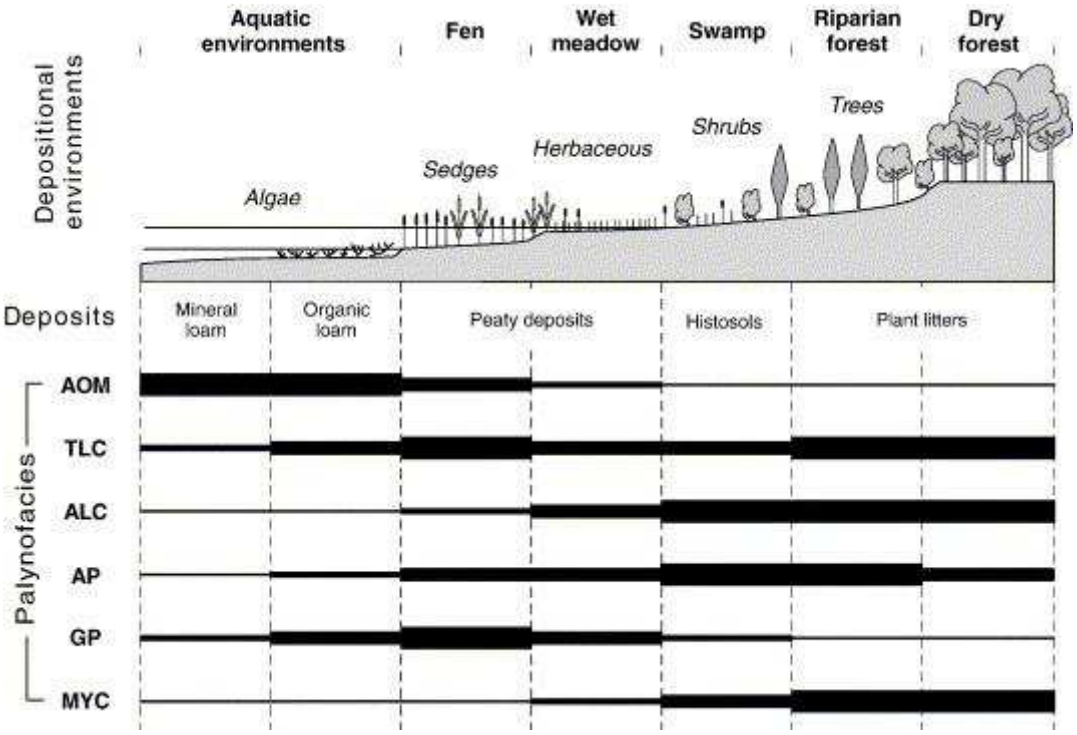

Figure 6: Palynofacies and depositional environments. Relationship between the depositional environments, the local vegetation, the nature of typical deposits and the composition of sedimentary OM (from Sebag, 2002).

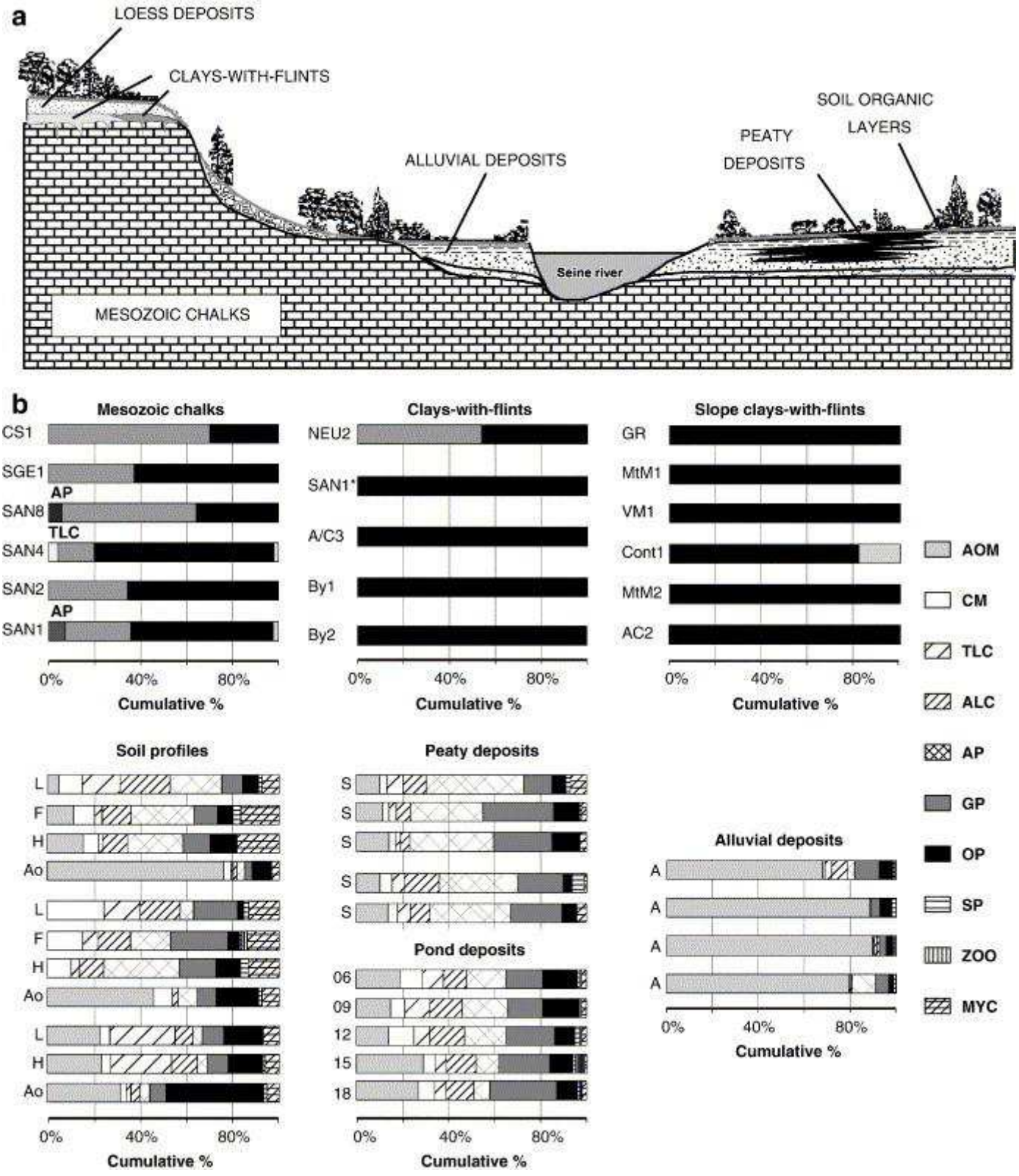

Figure 7: Distribution of palynofacies from the various compartments of the Lower Seine Valley (from Sebag et al., 2003).

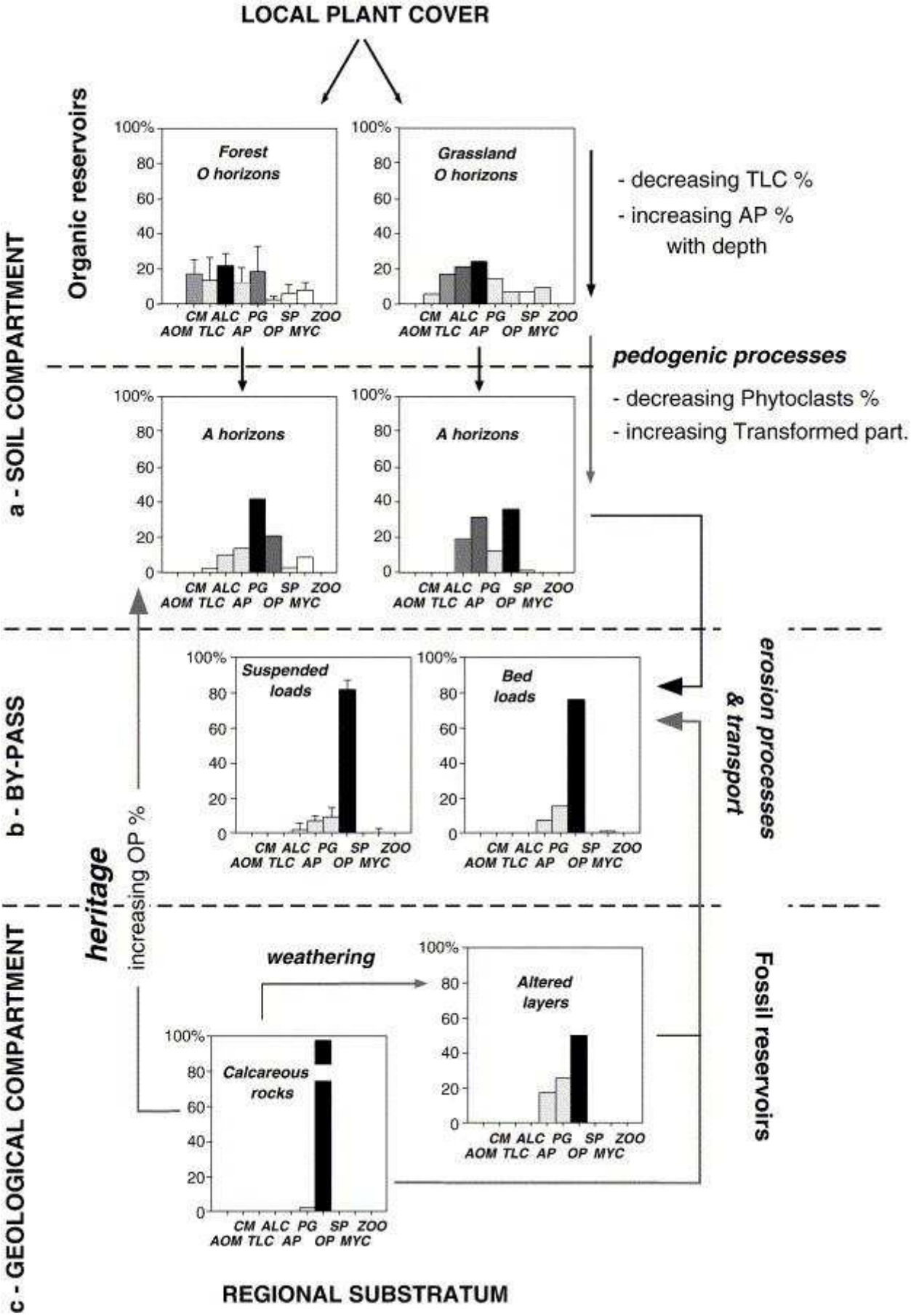
LOCAL PLANT COVER
Organic reservoirs
Forest
O horizons
Grassland
O horizons
- decreasing TLC %
- increasing AP %
with depth
pedogenic processes
- decreasing Phytoclasts %
- increasing Transformed part.
A horizons
A horizons
a - SOIL COMPARTMENT
b - BY-PASS
Suspended
loads
Bed
loads
erosion processes
& transport
heritage
increasing OP %
weathering
Altered
layers
Calcareous
rocks
Fossil reservoirs
c - GEOLOGICAL COMPARTMENT
REGIONAL SUBSTRATUM
100%
80
60
40
20
0
CM ALC PG SP ZOO
AOM TLC AP OP MYC

Figure 8: Overview of the particulate OM transfers in the Draix catchment (modified from Copard et al., 2006).

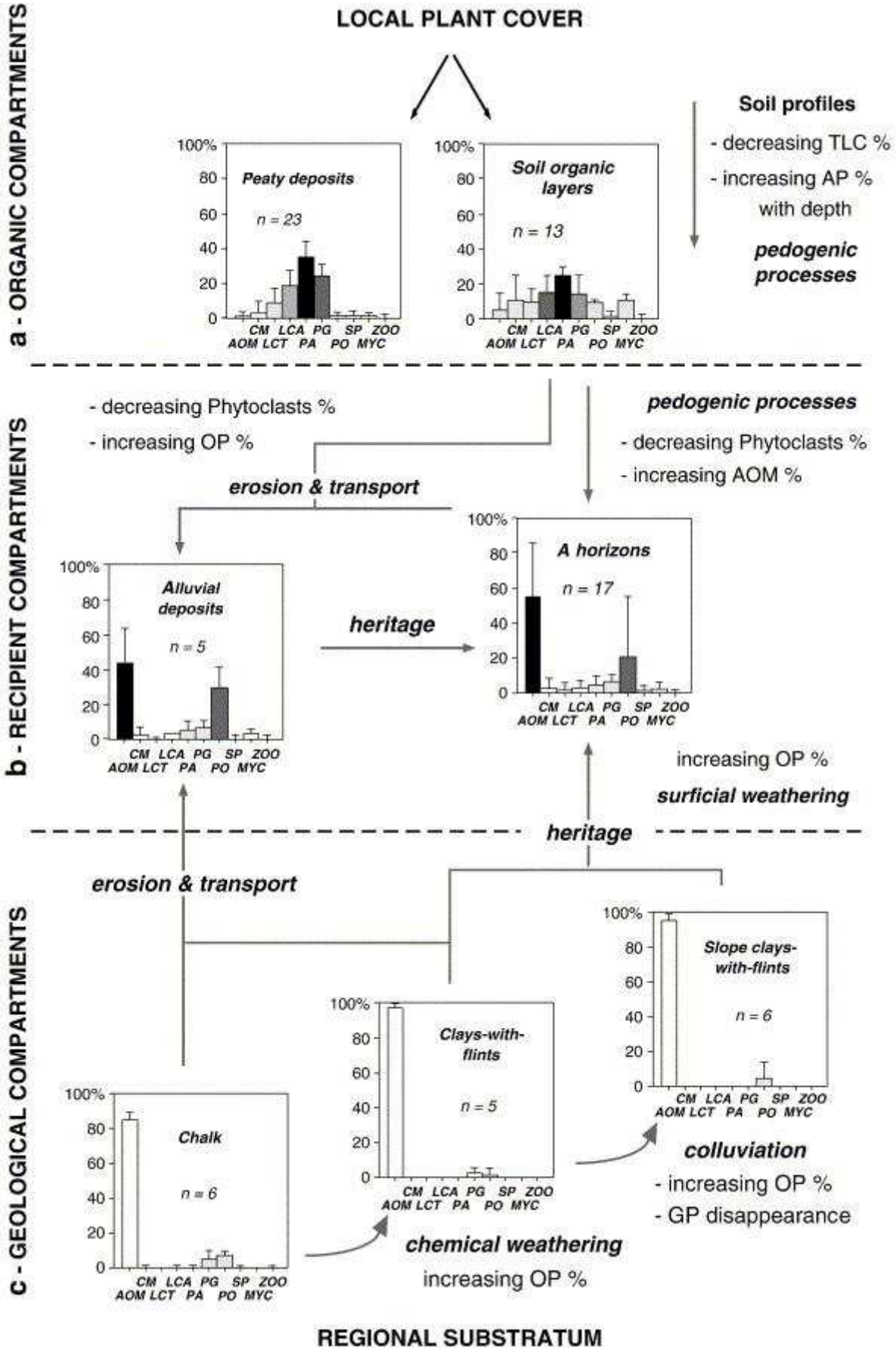
a - ORGANIC COMPARTMENTS
LOCAL PLANT COVER
Soil profiles
- decreasing TLC %
- increasing AP %
with depth
pedogenic processes
Peaty deposits
n = 23
Soil organic layers
n = 13
CM LCA PG SP ZOO
AOM LCT PA PO MYC
b - RECIPIENT COMPARTMENTS
- decreasing Phytoclasts %
- increasing OP %
erosion & transport
pedogenic processes
- decreasing Phytoclasts %
- increasing AOM %
Alluvial deposits
n = 5
heritage
A horizons
n = 17
increasing OP %
surficial weathering
heritage
erosion & transport
c - GEOLOGICAL COMPARTMENTS
Slope clays-with-flints
n = 6
Clays-with-flints
n = 5
Chalk
n = 6
colluviation
- increasing OP %
- GP disappearance
chemical weathering
increasing OP %
REGIONAL SUBSTRATUM

Figure 9: Overview of the particulate OM transfers in the Seine Estuary catchment (from Sebag et al., 2003; Laignel, 2003).