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# Recent hydrological variability of the Moroccan Middle-Atlas Mountains inferred from micro-scale sedimentological and geochemical analyses of lake sediments

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### Abstract

Study of the hydro-sedimentary dynamics of lakes provides key information on hydrological changes. In this work, we investigate Lake Azigza in the Moroccan Middle-Atlas, a region that suffers from a scarcity of observational hydrological data necessary for a coherent management of water resources. Sedimentary deposits of Lake Azigza (32°58'N, 5°26'W, 1,550 metres a.s.l.) were dated and analyzed by combining geochemical and mineralogical measurements coupled with microfacies characterization for the last 134 years. The detrital component derived from XRF-elemental composition and the microstructures observations of the lake sediments provided proxies of runoff activity and lake-level changes respectively. These proxies were calibrated with regional hydro-climatic and instrumental measurements available over the last 50 years and used to reconstruct past hydrological changes on interannual to decadal time-scales between 1879 and 2013. Since 1879, lake level and runoff proxies responded in phase to regional inter-annual precipitation variations. We also show that after the major lake-level drop observed in 2008, the response of the runoff proxy to variable precipitation regime is enhanced. Such an approach emphasizes the potential of these hydroclimate-sensitive sedimentary archives to assess the impact of climate change in the Mediterranean region.

Keywords: North Africa hydrology, Moroccan Middle-Atlas, lake sediments, microfacies, XRF, runoff, lake level

### Introduction

Global warming is expected to increase the frequency and/or intensity of extreme precipitation and drought events (IPCC, 2013), particularly in the Mediterranean basin including Morocco (Gao et al., 2006; Giorgi and Lionello, 2008; Tramblay et al, 2012). These variable hydrological conditions could significantly impact the water resources in the Moroccan Middle-Atlas region (Bouaicha and Benabdelfadel, 2010). For Morocco, this hypothesis has been tested for different climate scenarios. Using a set of regional climate models with a variable-resolution configuration, Driouech et al. (2010) have forecasted a decrease in mean precipitation over Morocco associated with changes in the distribution and intensification of extreme events for the period 2021–2050. The maximum drought duration is estimated to increase over most of the country, especially in the western regions of the Atlas Mountains (Driouech et al., 2010). The climate of the Middle-Atlas region is influenced by air masses coming from the Mediterranean, Saharan desert and Atlantic Ocean (Knippertz et al., 2003; Ouda et al. 2005). Combined with a steep orography, this climate setting leads to a precipitation distribution characterized by high spatial variability, a pronounced seasonality and a strong inter-annual variability (Ouda et al. 2005). Most of the meteorological observations are only available for the last 40 years, based on a station network particularly sparse in the Middle-Atlas region (Driouech et al., 2010; Tramblay et al., 2012; 2013). In this region, it is a key challenge to extend the hydro-climate dataset beyond the instrumental record period in order to understand the full spectrum of past hydrological variability.

Analysis of natural archives such as lake sediments can potentially give access to the preinstrumental hydro-climate data. Several studies have already shown the usefulness of lake sediments in reconstructing paleohydrological conditions in association with rainfall-induced runoff (Czymzik et al., 2013; Gilli et al., 2013). Sedimentological and geochemical analysis on the sedimentary sequences can provide past reconstructions of flood frequency (Lapointe et al., 2012; Wilhelm et al., 2012; 2014; Giguet-Covex et al., 2012; Czymzik et al., 2013; Vanniere et al., 2013) as well as soil erosion (Simonneau et al., 2013). Corella et al. (2014) compared daily maximum precipitation with detrital microfacies observed in lacustrine sequences (thickness and occurrence) for the period 1917-1994 and reconstructed annual floods events recorded since the 14th century in the North-Eastern Iberian Peninsula. Such an approach derived from lacustrine paleoflood records calibrated with instrumental data can substantially improve the quality of long-term flood reconstructions (Wilhelm and Ballesteros-Cànovas, 2016). However, several issues remain on the understanding of hydrosedimentary dynamics and weaken the reliability of such records. The response of shallow lakes to hydrological variability modulates their water levels and impacts the amount and/or source of clastic materials filling the lake. Indeed, changes in the source-to-sink relationship also depend on the geomorphological setting of the watershed and should be taken into account for the reconstruction of hydrological events.

The Middle-Atlas mountains are considered as the "Moroccan water tower", with its catchment feeding the two most important rivers of the country: Oued Oum-Er-Rbia (555 km long) and Oued Sebou (456 km long), both flowing into the Atlantic Ocean. The region contains several natural lakes lacking any surface outlet, which act as natural pluviometers very sensitive to hydro-climatic variations (Benkaddour 1993; Damnati, 2000; 2009). For example, sedimentological, geochemical and mineralogical analyses of the Lake Iffrah sediments, a tectono-karstic lake situated ~80 km North-East of Lake Azigza, reveal a lake-level drop interpreted as the result of successive dry periods over the past three decades, and

eutrophication linked to human activity (Etebaai et al., 2012). Similar trends have been recorded in other lacustrine sediments of the Middle-Atlas covering the same time interval (Damnati et al., 2012). Unfortunately, these studies are not based on the calibration of sedimentological proxies with hydro-climate data, and the lacustrine sequences are often subject to dating problems and low temporal resolution of the data.

This study is focused on short sediments cores retrieved in the deep basin from Lake Azigza in the Middle-Atlas in 2013. Analyses of the elemental composition (X-ray fluorescence: XRF), microstructure, mineralogy, granulometry and radionuclides dating of the sedimentary sequences were performed. These data combined with the geomorphological properties of Lake Azigza were used to identify relevant proxies of lake water level and runoff activity. For the interval dated with radionuclides (1963-2013), the sedimentary proxies are calibrated using regional hydro-climate data. Finally, the runoff activity and lake-level proxies are applied to the entire sequence to infer past hydrological changes over the last 134 years.

### **Study site**

The Moroccan Middle-Atlas is an intracontinental mountain range, which exhibits a large synclinal trough extending 350 km along a NE-SW oriented belt. The geology of the region is characterized by tectono-karstic systems, represented by calcareous dolomites deposited during the Early Jurassic, overlying Triassic mudstones (Martin, 1981). Because of its high elevation and the influence of oceanic precipitation, the Middle-Atlas has relatively cold and humid conditions and is considered as a mountainous Mediterranean climate area with mean annual temperatures of about 12°C, and an annual precipitation of about 900 mm (Martin, 1981).

Lake Azigza ( $32^{\circ}58^{\circ}N$ ,  $5^{\circ}26^{\circ}W$ ) is an endorheic and monomictic lake with tectono-karstic origin (Fig. 1A, B), located in the Moroccan Middle-Atlas (1550 m a.s.l). Nowadays, the lake has no surface outflow and occupies a unique depression with an area of about 0.55 km<sup>2</sup> (measured in April, 2013), having a maximum water depth of 42 m and a mean depth of 26 m (Fig. 1). Its catchment area is about 10.2 km<sup>2</sup>, covered by a green oak forest and a cedar forest in the western part of the basin associated with the development of brunified fersialitic red soils (Flower et al., 1989). These authors (*op. cit.*) also noted the low level of human disturbance in the watershed. The climate in this region is of Mediterranean sub-humid type, characterized by wet winters and dry summers with mean annual rainfall (MAP) of 900 mm (Martin, 1981).

The region is however characterized by a strong inter-annual variability. For example, measurements obtained from a 4-year meteorological site monitoring at Lake Azigza show that MAP is about 690 mm (Vidal et al., 2016). Over the past decades (1979-2015), MAP can differ by as much as 80% from the mean value (Vidal et al., 2016). Hydrological results from Lake Azigza indicate that water levels are linked to the precipitation regime (Vidal et al., 2016), a results that has been confirmed using hydrological mass balance modelling applied to Lake Azigza (over the last 4 years) (Adallal et al., submitted).

Previous studies have been conducted on the physico-chemical and limnological properties of Lake Azigza (Gayral and Panouse, 1954; Flower et al., 1989; Flower and Foster, 1992; Benkaddour, 2008). Observations of paleoshorelines in the catchment indicate that plurimetric changes in lake level have occurred during the past few decades (Flower and Foster, 1992).

### Materials and methods

#### Materials

Three short cores AZA13-1 (64.5 cm), AZA13-3 (80.6 cm) and AZA13-4 (16 cm), were recovered in April 2013 in water depths of 30.3, 32.1 and 16.4 m, respectively, using a UWITEC gravity corer (63 mm diameter) (Fig. 1E and F). High-resolution bathymetric profiles were performed using a Humminbird 898c SI (83 kHz; bathymetry shown in Fig. 1F). All analyses were carried out at CEREGE (Aix-en-Provence), but some thin-sections were prepared at EPOC (Bordeaux) and radionuclide measurements were performed at Géosciences (Montpellier).

#### **Elemental composition and radiography**

The ITRAX Core Scanner (Cox Analytical Systems) allows the simultaneous high-resolution acquisition of chemical composition of the sample by XRF scanning as well as the micro-variations in density and structure (microradiography) (Croudace et al. 2006). The positive X-ray signal obtained by radiography means that the darker the image the denser the sediment (Croudace et al., 2006). Analyses of K, Si, Ca, Ti, Fe, Sr and Mn, as well as the inc/coh (incoherent/coherent) ratio were performed on cores AZA-13-1 and AZA-13-3 with a 1 mm resolution and 15 s exposure time, using a Mo X-ray tube operating at 30 kV and 25 mA. The inc/coh value corresponds to the Compton to Rayleigh scattering ratio, representing the relative abundance between light and heavy elements (Croudace et al. 2006; Chawchai et al., 2016). The potential matrix effect on elemental XRF intensity was tested by comparing raw XRF elemental profiles with XRF intensity normalized against total counts. Since the main

features of the elemental profiles are insensitive to normalization, we present elemental XRF intensity without the normalization.

### Mineralogy

The mineralogical composition of the sediment was determined with an X-ray  $\theta$ - $\theta$  diffractometer (X'Pert Pro MPD, Panalytical) using Co K $\alpha$  radiation ( $\lambda$ = 1.79 Å), operated at 40 kV and 40 mA. Mineral phases were identified using the International Center of Diffraction Data PDF-2 database under the X'Pert Highscore plus software (Panalytical). Six samples (10 cm step) from core AZA-13-1 were ground in an agate mortar, providing powders that were placed on monocrystalline silicon plates cut parallel to the (501) face to ensure a very low background and absence of interfering diffraction peaks. These zerobackground silicon plates were prepared with a droplet of ethanol to obtain a thin and homogeneous layer of powder. Statistical precision was improved by spinning the samples at 1500 rpm. A counting time of 22 s per 0.033° step was applied over a range 5–80° 2 $\theta$ .

Clay minerals were separated from six bulk samples (at 10 cm intervals) taken from core AZA-13-1 using a sedimentation method according to Stokes' law, after removing carbonates and organic matter with HCl (2M) and hydrogen peroxide (30% vol.), respectively. After ultrasonic disaggregation, the clay suspension was then deposited onto three glass slides to prepare oriented samples (for reference, ethylene-glycol saturation and heat treatment). Ethylene-glycol saturation was carried out on the oriented sample at ambient temperature in a glass desiccator to identify swelling clays (smectite or mixed-layer minerals containing smectite). The samples were heated in an oven at 490°C for four hours to distinguish kaolinite from chlorite.

### Total organic carbon (TOC%) and Total inorganic carbon (CaCO<sub>3</sub>%)

The total carbon (TC%) and total organic carbon (TOC%) contents were determined with a FISONS NA 1500 elemental analyzer. The protocol applied in this study follows the procedure of Pailler and Bard (2002). Six samples (10 cm step) from core AZA-13-1 were desiccated in an oven for four days, and then ground and homogenized in an agate mortar. TOC% was measured after acid digestion to remove the carbonate fraction, and each sample was duplicated. Calcium carbonate contents were calculated using the following equation:

 $CaCO_3$  (%) = (TC-TOC) × 8.33

#### Laser diffraction grain size

The 14 samples (5-cm step) from core AZA-13-1 were fine grained, with a general grain size  $< 63 \ \mu\text{m}$ . Organic matter was removed prior to analysis with hydrogen peroxide and dispersed using 0.3% sodium hexametaphosphate. The grain-size distribution was measured using a Beckman Coulter LS 13 320 laser granulometer with a range of 0.04 to 2000  $\mu$ m. The calculation model uses the Fraunhöfer and Mie theories. All samples containing fine particles were thus measured at obscuration levels between 8 and 12% and between 45 and 70% with PIDS (Polarization Intensity Differential Scattering). All samples show a bimodal grain size distribution with modes centred on clays and silts (Supplementary Figure 1). In the following, results are shown for the clay fraction (< 3  $\mu$ m) and silt fraction (>3  $\mu$ m).

#### Microfacies

#### Thin-section preparation

For core AZA-13-1, continuous thin sections were performed at EPOC. Sediments were subsampled using perforated aluminum slabs, before being dehydrated using the acetone-water exchange technique (Bénard, 1996; Zaragosi et al., 2006) and impregnated with an epoxy resin. After polymerization, resin-impregnated blocks were cut to obtain thin-sections 10 cm in length, 4 cm in width and 30 µm thick using a rotating lapidary unit.

For core AZA-13-4, thin sections were obtained at CEREGE. Sediments were subsampled with aluminum slabs and immersed in liquid nitrogen, before being lyophilized using the Lyophilisator CHRIST Alpha 1-2 LD plus. They were then impregnated with Araldite 2020 epoxy resin. Impregnated blocks were then cut to obtain sections 4.5 cm in length, 2.5 cm in width and 30 µm thick using a rotating lapidary unit.

#### Microfacies characterization

The thin-sections with dimensions of 10 cm x 4.5 cm were scanned using the EPSON 3200 PHOTO before analysis of microfacies under a Leica DM6000B instrument. Full motorization of the microscope stage (in X, Y and Z) allows acquisition of a mosaic of several cm<sup>2</sup>. Microfacies were also analyzed by energy dispersive spectroscopy (EDS). At the same time, a Si(Li) detector microprobe installed on the Hitachi S-3000N scanning electron microscope (SEM) was used for semi-quantitative elemental analysis. The generator was set at 15 kV, with an EDS acquisition time of 90 min. Elemental mapping (Si, Ca, Fe or Si, Ca and K) of grains in the microfacies allows comparisons between the mineralogy and particle size of the sediment.

#### **Age-depth model**

Radionuclide activities of bulk sediment were measured on 13 samples from core AZA-13-3 between the top and 39 cm down core. Dating of sedimentary layers was carried out using <sup>210</sup>Pb and <sup>137</sup>Cs. These nuclides, together with U, Th, and <sup>226</sup>Ra, were determined by gamma spectrometry at Géosciences Montpellier. Sediment subsamples from 1-cm-thick intervals were crushed after drying, transferred into small gas-tight polyethylene terephthalate tubes, and then stored for more than three weeks to ensure equilibrium between <sup>226</sup>Ra and <sup>222</sup>Rn. The activities of the nuclides of interest were measured using a Canberra Ge well detector and compared with the known activities of an in-house standard. Activities of <sup>210</sup>Pb are obtained by integrating the area of the 46.5-keV photo-peak.<sup>226</sup>Ra activities are determined from the average of values derived from the 186.2-keV peak of <sup>226</sup>Ra and the peaks of its daughter products in secular equilibrium with <sup>214</sup>Pb (295 and 352 keV) and <sup>214</sup>Bi (609 keV). In each sample, the <sup>210</sup>Pb excess activities (<sup>210</sup>Pb<sub>ex</sub>) are calculated by subtracting the <sup>226</sup>Ra supported activity from the total <sup>210</sup>Pb activity.

### Results

#### Age model

In core AZA-13-3, the <sup>137</sup>Cs activity significantly increases at about 27 cm depth (Fig. 2). The Cs peak (200 mBq/g) is due to atmospheric nuclear tests and the associated radioactive fallout

between 1962 and 1964 (Cambray et al., 1989). Based on this peak event, a mean sedimentation rate of core AZA-13-3 is estimated at 0.54 cm.year<sup>-1</sup>. The gradual down-core decrease of the <sup>210</sup>Pb<sub>ex</sub> activity (for the first 39 cm) suggests that the radionuclide signal is not affected by bioturbation (Fig. 2A). The Constant Flux/Constant Sedimentation (CFCS) model applied to the <sup>210</sup>Pb<sub>ex</sub> data yields a mean sedimentation rate of about 0.53 cm.year<sup>-1</sup>. Since similar results are obtained from <sup>210</sup>Pb<sub>ex</sub> and <sup>137</sup>Cs, we assume a constant sedimentation rate of 0.54 cm.year<sup>-1</sup> during the last 50 years (at 27 cm depth in core). The assumption of constant sedimentation rate is supported by the absence of drastic changes in grain size (clay, silt and sand) along the entire core AZA-13-3 (Fig. 3 and Supplementary Figure 1).

The age-depth model for core AZA-13-3 was applied to core AZA-13-1 using the Analyseries software to correlate K-XRF intensities measured on both cores (Paillard et al., 1996; Fig. 2B). The correlation points based on the K-XRF profiles indicate a good correspondence between both records for the first 28 cm of the cores and a slight sediment compaction in core AZA13-1 after 28 cm to the base of the sequence (Fig. 2B). The age model, obtained considering a linear interpolation between these points, shows that core AZA-13-1 covers the last 134 years (Fig. 2C), with a yearly (even slightly less) temporal resolution considering the XRF records.

#### Sedimentology

The lacustrine sedimentary facies are classified according to Schnurrenberger et al. (2003). The sedimentary sequences are composed of light-brown to dark, unconsolidated, matrixsupported, partly laminated clastic sediments and autochthonous carbonates. The term autochthonous carbonate refers to primary inorganic precipitation in the water column and post-depositional authigenic precipitation at the water-sediment interface and within sediment pore waters.

#### Core AZA-13-1

In core AZA-13-1, three facies are distinguished based on photographs, radiography (Fig. 3) and petrographic/SEM-EDS analyses (Fig. 4). Facies 1 is found at 64.5-55 cm and 29-16 cm, and is composed of light-brown sediments (Fig. 3) with several sub-horizontal discontinuous and mm-thick laminations (Fig. 4A). These laminations contain wood fragments, as shown by the presence of epithelial cells (Fig. 4A). Calcareous shells of ostracods and other bivalves are also present, as revealed by the SEM-EDS results (Fig. 4A). This facies is characterized by a high sediment density (Fig. 3, radiography image) and low inc/coh values (Fig. 3). Facies 2 is present within two depth intervals (55-29 and 16-5 cm; Fig. 3) and consists of homogeneous light-brown sediments composed of mm-sized bivalve and ostracod shells (Fig. 4B). The absence of laminations containing wood fragments is the main difference with Facies 1 (Fig. 4A and 4B). Also, the frequency of laminated layers in the down-core records varies between the different facies, with seven occurrences for type 1 called type-1 microstructures (T1M) against only three for type 2 (T2M) (Fig. 4A and 4B). Facies 3 occurs only at the top of the core (between 1 and 5 cm; Fig. 3). It displays laminations, corresponding to the type 3 (T3M) with alternations of autochthonous carbonate layers and quartz-rich sediments (Fig. 4C).

Grain-size analyses of the total fraction of the sediment show that the sediment is mainly represented by silts (mean relative abundance of 73.5%), a result which is compatible with the trends observed in CaCO<sub>3</sub>% and Ca-XRF except in the basal part (Fig. 3). Carbonates are mainly calcite, with small proportions of dolomite and aragonite (Supplementary Figure 2). We consider that early diagenesis leading to calcite precipitation is negligible since no sparitic

cement is observed in either the petrographic or EDS analyses. The clay fraction (mean relative abundance of 26.5%) evolve in parallel with the contents of K, Ti, Fe and Si (Fig. 3), and is mainly represented by interstratified clays, smectite, illite and kaolinite (Supplementary Figure 3).

#### Core AZA-13-4

Because of their proximity to the shoreline, the sediments in core AZA-13-4 are coarser than in core AZA-13-1 (Fig. 5). AZA-13-4 is a short core containing brown unlaminated sediments with a coarse layer in the middle of the sequence. SEM analyses show that the sediment is mainly composed of detrital Si-rich minerals, of sand to fine gravel grain size, with wellpreserved or fragmented millimetric calcareous shells of ostracods and other bivalves (Fig. 5).

#### Geochemistry

In core AZA-13-1, XRF profiles show a co-variation between Fe and Ti ( $R^2=0.81$ ; Table 1A) in close association with K (Fig. 3). The K and Si profiles are also strongly correlated ( $R^2=0.74$ ; Table 1A) and show a similar variability with the clay fraction (Fig. 3). The inc/coh profile is almost a mirror image of the profiles of detrital elements (K, Si, Fe and Ti) (Fig. 3), displaying a co-variation with TOC% values except at the bottom of the core (Fig. 3). This may be due to the highly heterogeneous distribution of wood fragments in the sediment (Fig. 4A). The Ca contents co-vary with CaCO<sub>3</sub>% and the Sr/Ti ratio (Fig. 3). In general, Sr is enriched in aragonite, but the detrital fraction also contains terrigenous Sr. Similar depth-profiles of Ca, CaCO<sub>3</sub>% and Sr/Ti are consistent with the mineralogical results indicating the presence of calcite and aragonite (Supplementary Figure 2). Apart from the interval between 64.5 and 48 cm, the carbonate phases show similar profiles compared with silt (Fig. 3).

Principal Component Analysis (PCA) is applied here to K, Si, Ca, Ti, Fe, Mn, Sr and the inc/ coh ratio. Elemental data is first centred to zero by subtraction of averages and then scaled with variance to give equal weight to each element (Weltje and Tjallingii, 2008). PCA yields two main factorial axes (PC1 and PC2) which represent about 76.03% of the entire variability and which allow us to distinguish three chemical end-members (Table 1A and Supplementary Figure 4A). PC1 is controlled by variations in the detrital elements Fe, Ti, Mn, K, Si and Sr (first chemical end-member) and Ca (second chemical end-member). PC2 is controlled by Ca, which is itself anti-correlated with the inc/coh ratio (third chemical end-member); this ratio is a proxy of organic matter and water content in the sediment (Burnett et al., 2011; Guyard et al., 2007; Jouve et al., 2013). The correlation between Sr/Ti and Ca implies that carbonates primarily result from autochthonous precipitation. SEM-EDS analyses on microstructures type 1, 2 and 3 also support that carbonates are mainly originated from biogenic and endogenic precipitation (Fig. 4). The occurrence of dolomite indicates that minor contribution from detrital carbonates cannot be excluded (Supplementary Figure 2). The relatively low Rock-Eval hydrogen index values (HI = 310 mgHC/g TOC) (Supplementary Table 1) in all samples suggest that the organic matter originates from a mixture of land-plant and algal production, and subsequently escaped remineralization during sedimentation (Carroll, 1998; Meyers, 2003). Organic matter is thus mainly derived from the catchment area (allochthonous). As a result, the opposite trend between Ca and inc/coh also supports the hypothesis that carbonates are mainly derived from autochthonous precipitation.

The relationships between the two main factorial axes of the PCA are compared with the identified sedimentary facies described previously. Facies 1 is characterized by a positive correlation between PC1 and PC2 (Supplementary Figure 4B), clay fraction and CaCO<sub>3</sub> content (Fig. 3). The inc/coh ratio decreases in this facies together with TOC values. The TOC

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content remains high at the base of the core due to the particularly high abundance of wood in the sediment. The geochemical signature of Facies 2 is characterized by a positive correlation between PC1 and PC2 (Supplementary Figure 4B), associated with lower PC1, PC2, clay fraction and CaCO<sub>3</sub> values (Fig. 3) and typically shows higher inc/coh ratio and TOC values. Facies 3 shows a negative correlation between PC1 and PC2 (Supplementary Figure 4B), but much higher PC1 and clay values, and lower CaCO<sub>3</sub> and TOC values (Fig. 3).

### Discussion

Changes in lake level and runoff activity both have impacts on the source-to-sink processes leading to infilling of the lake. Geochemical, micro-structural and mineralogical analyses of the sediment provide crucial information to understand the link between hydro-sedimentary dynamics of the lake and precipitation regime.

In the following, we propose a proxy for runoff activity inferred from a combination of detrital elements (based on XRF intensity data) and a lake-level proxy derived from microstructural analyses. To assess the reliability of the proposed hydro-sedimentary indicators, the lake water level and runoff proxies are calibrated using regional hydro-climate data covering the past 50 years corresponding to the interval dated with radionuclides (1963-2013). Then, we extend our approach to the entire sedimentary sequence, thus providing a hydrological reconstruction over the last century for the Middle-Atlas region.

#### Hydro-sedimentary processes

#### **Proxy of runoff activity**

The presence of Fe, Ti, K, and Si-rich particles in core AZA13-1 is mainly associated with silicate minerals and clearly reflects the PC1 factorial axis (Table 1B). We therefore consider PC1 as an indicator of the detrital fraction of Lake Azigza sediments, which is closely linked to runoff activity in the catchment.

PC1 is related to quartz, kaolinite, illite, interstratified clays (probably illite/smectite) and chlorite (Supplementary Figures 2 and 3). Detrital quartz grains are likely derived from sandsized and altered Si-rich minerals detected in cores AZA-13-4 (core closer to the shoreline, Fig. 5), which are probably derived from Triassic sandstones (Martin, 1981; Benkaddour, 1993). Potassium has been used as a proxy for clay minerals derived from soil weathering and erosion (Unkel, 2008; Cuven et al., 2010; Kylander et al., 2011; Hahn et al., 2014). This element is also associated with illite and chlorite minerals (Deer et al., 1992; Minyuk et al., 2007) that are detected in Lake Azigza sediments (Supplementary Figure 3). The similar trends shown by clays, K and Ti indicates that the clay fraction is mainly derived from detrital inputs, and not from post-depositional processes, since Ti is mainly present in insoluble detrital particles (Demory et al., 2005; Jouve et al., 2013; Hahn et al., 2014). The presence of detrital minerals in Lake Azigza could also be linked to aeolian processes in addition to runoff activity since mass movement deposits are absent within the sediments. Microscopic observation of the detrital silt fraction reveals angular quartz grains (see an example in facies 3, Fig. 4C). This feature rules out the possibility of distal wind transport. Local winds could also transport detrital particles to the deep basin. However, at Lake Azigza, the wind intensity (~max. 4.3 km.h<sup>-1</sup>) is probably not strong enough to mobilize a significant proportion of the

detrital particles to the deep basin. Moreover, the silt fraction of the sediment is easily mobilized and maintained in suspension by the action of the wind (Tucker, 1991). In any case, the silt size fraction is mainly represented by autochthonous carbonates and not by detrital particles. Thus, we consider that the transport of clays would have been preferentially induced by runoff. The occurrence of small gullies (observed in the proximal catchment, Fig. 1D) incised into kaolinite-rich soils may have facilitated the mobilization of detrital clay particles during runoff activity.

The main hydro-sedimentary process influencing the detrital sedimentation pattern can be described in two steps. First, increased precipitation causes a rapid rise in lake level (confirmed by measurements from a submerged probe and the meteorological station at Lake Azigza; Vidal et al., 2016). Second, the ensuing runoff carries more detrital particles with finer grain size, due to increased distal source materials and decreased availability of soil particles with the proximity of the forest. Other factors related to watershed-lake processes linking soil and vegetation dynamics may also influence detrital material available for the runoff. Rock-eval HI values and mineralogical composition of the deep basin sediments suggest similar vegetation and soil conditions during the last century (Supplementary-Figure-2). Aerial photographs of Lake Azigza over the past decades (1960s, 1970s and 2000s; Figure 8) do not reveal any drastic change in the cedar forest extent. Taken together, the available observations at Lake Azigza suggest that watershed conditions remained relatively stable over the study period with negligible impact on the runoff activity.

#### Proxy of lake-level change

In core AZA13-1, micro-scale sedimentological analyses reveal two types of structures resulting from a high hydro-dynamic regime characterized by discontinuities and biological

components derived from the shorelines (Fig. 4A). The first type (T1M, present in Facies 1) is characterized by fragments of ostracods, bivalves and wood, while the second type (T2M, present in Facies 2) differs from the previous one by the absence of wood.

The basal coarse sand layers, associated with a fining-upward intermediate sequence and a top clay cap, are typical structures derived from hyperpycnal flows generated by floods, and have been described in several lakes in various geomorphological settings (Mulder et al., 2001; 2003; St Onge et al., 2004; Schneider et al., 2004; Chapron et al., 2006; Guyard et al., 2007; Wilhelm et al., 2012; 2013; Corella et al., 2014; Jouve et al., 2017). On the other hand, in their study of a shallow lake (30 m depth) with steep slopes, Corella et al. (2014) were able to differentiate flood-induced turbidites from microstructures formed under lower energy flow conditions. Turbidites and graded beds are associated with rainfall events under higher energy hydro-dynamic conditions than during the formation of discontinuous detrital layers.

In the Lake Azigza deep basin cores, there is no evidence for graded beds that could be related to flood-induced turbidites. Hence, the sedimentary structures are composed of different shoreline materials without any drastic change in grain size (Fig. 3 and Supplementary Figure 1). This feature cannot be explained solely as being due to transport by low-density currents after a rainfall event. We need to consider the distance between the downstream gullies and the forest edge, which probably modulates the presence of wood fragments on the shoreline and their transport towards the deep basin. Wood fragments and biogenic carbonates (originating from the shoreline area) are present in Facies 1 (Fig. 4A). Facies 1 is also characterized by a high content of detrital proxies (PC1) and high sediment density (Fig. 3). During high lake level stands, the shorelines are closer to the cedar forest, so the steeper slopes (Fig. 1C) increase the intensity of surface runoff in the lakeshore area and enhance transport to the deep basin. Consequently, the sedimentological and geochemical features of

Facies 1 are interpreted as indicators of high lake level stands. This observation is further supported by the abundance of diatoms and the composition of the assemblages, dominated by *Cyclotella atomus* and *Cyclotella stelligera*, which are indicators of freshwater inputs (Supplementary Figure 5).

Facies 2 shows sedimentary structures containing ostracods and other bivalve shells, but *without* wood fragments (Fig. 4B), i.e. these sediments have lower density and less abundant detrital minerals (Fig. 3). A reduced transport of detrital minerals to the deep basin is consistent with a decrease in runoff activity and can be linked to gentler slopes (Fig. 1C). In contrast to the interpretation developed for the high-energy sedimentary microstructures, Facies 2 can be related to low lake level conditions. This is consistent with a decrease in the detrital fraction and a relative increase in organic matter (Fig. 3). In addition, Facies 2 is characterized by a lower abundance of frustules and/or assemblages dominated by *Cyclotella spp.*, associated with benthic species (Supplementary Figure 5).

#### **Calibration of the proxies**

We now compare the geochemical signals with hydrological data. We use regional precipitation observations obtained from the Climate Research Unit (CRU) database. The geo-referenced CRU TS 3.22 dataset provides monthly precipitation on a grid with a spatial resolution of 0.5° in latitude and longitude centred at 5.75°W; 32.75°N for the period 1901 to 2013. This dataset compares well with the inter-annual variability of local precipitation, even if the monthly amount of precipitation is underestimated in the re-analysis of the data (Adallal et al., submitted).

As a first step, we consider only annual precipitation from the CRU-database for the period 1963-2013, which corresponds to the dated interval of the Azigza sedimentary sequences

(Fig. 6). Annual PC1 and K values are well correlated with annual precipitation (R=0.59 and R=0.54, respectively) for the period 1963-1999 (Fig. 7A). During the 1960s and 1970s, precipitation is significantly above average values. This period seems to be associated with relatively high runoff activity (PC1). The increased runoff enhances the detrital input, favouring the presence of autochthonous carbonates and diluting organic matter in the sediment. During the 1980s and 1990s, precipitation decreases as well as runoff activity and autochthonous carbonates, counter-balanced by an increase of organic content in the sediment (Fig. 3). From 2000 to 2008, the runoff signal rises drastically. This marked trend is accompanied by a short-term increase in monthly precipitation (Fig. 6).

Our reconstruction is also consistent with lake-level indicators obtained from on-site measurements, historical aerial photographs and earlier studies (Gayral and Panouse, 1954; Flower et al., 1989; Flower and Foster, 1992) (Figs. 6 and 8). For example, the aerial photography from 1964 provides evidence of a high lake level, about 10 m above the 2013 level (our reference level) (Fig. 8). During this period, the sediment is classified into Facies 1 corresponding to a high lake level stand with increased runoff activity (Fig. 6). Flower and Foster (1992) estimated lake level changes using the position of the shorelines between 1979 and 1984, as well as in 1990 (but no photographs are available). Compared to our reference level for 2013, we estimate that the lake level was about 3 m lower in 1990. Facies 2 is developed in the sedimentary sequence deposited at that time (Fig. 6), compatible with a low lake-level stand with a smaller distance between the coring site and the shorelines favouring deposition of coarser calcareous shells to the deep basin during rainfall events. The same observation can be made for the interval deposited in 2008, which records a major lake level drop (Fig. 6, and 8C).

The high-resolution lake bathymetry clearly documents the hydro-sedimentary dynamics associated with low lake levels and the occurrence of paleo-channels in the NW and SE parts of the deep basin (Fig. 1F). Isobath structures highlight three submerged channels (grey arrows in Fig. 1F) situated between 0 and 25 m depth, which were probably reactivated during the recent 2008 lake-level drop. Two deeper and larger submerged channels (dotted grey arrows in Fig. 1F) between 20 and 30 m depth, provide evidence of past lake level drop(s) of much greater amplitude than in 2008.

At the top of the sequence, T3M correspond to the period shortly following the drastic drop in lake level in 2008 (about 7 m, Figs. 6 and 8). This drop is consistent with a decrease in runoff activity and precipitation during the 1990s and early 2000s (Fig. 6). Laminae of autochthonous carbonates are intercalated with laminae of coarser detrital grains (Fig. 4C), and are interpreted as annual laminated deposits (varves). The presence of autochthonous carbonates in the sediment is no longer linked to the runoff, but appears to be associated with supersaturated conditions in the water column during the spring/summer. Facies 3 has a geochemical signal marked by an anti-correlation between PC1 and PC2 (mostly Ca) (R=-0.77; Supplementary Figure 4B), which indicates hydro-climate conditions are recorded differently in lake sediments during the 2000s. Facies 3 is coeval with a strong increase in PC1 index (Fig. 6). Nevertheless, the interval (2000-2013) is still characterised by a correlation between PC1 and precipitation (R=0.6) but with an increase in the slope of the regression line (Fig. 7). The runoff proxy is linked to the availability of detrital materials due to enhanced erosional potential combined with drier conditions. The lake level decrease of 8 meters between 1979 and 1984 (Fig. 6), was resulting in an increase of erodible surface area that could supply the lake with detrital materials. In a context of low lake level conditions, the sensitivity of the proxy to precipitation variability is enhanced. The drastic lake level drop is

also consistent with the presence of a quasi-monospecific diatom assemblage dominated by *Cyclotella ocellata*, a species that is potentially very well adapted in the context of a nutrient-depleted water column (Supplementary Figure 5).

The Ca record is following the PC1 index. Hence, PC1 is conditioned by two chemical endmembers (detrital inputs and autochthonous carbonates; Supplementary Figure 4A) that are linked. In shallow waters close to the lake shore, supersaturated conditions systematically lead to the precipitation of autochthonous carbonates (aragonite and calcite; Supplementary Figure 2 and Benkaddour et al., 1993; 2008). During rainfall events, autochthonous carbonates present in the drawdown zone and detrital carbonates from the watershed can be mobilized by the runoff and supplied to the deep basin. The more the runoff, the higher the carbonates in lake sediments. On the other hand, the increased influx of Ca<sup>2+</sup> ions and nutrients due to higher surface runoff could favour the precipitation of autochthonous carbonates, as suggested by Brauer et al. (2008). While autochthonous carbonates in lacustrine sediments are generally used as a proxy of drier conditions (e.g. Damnati et al., 2016) and warmer climate (Meyers, 2003), at Lake Azigza, carbonates are primarily related to higher runoff activity under wetter conditions. However, when the response of the runoff proxy is enhanced due to increased erodible surface in the 2000s, Ca is diluted by the large amount of the detrital elements (Ti, Fe, Si, Sr, Mn and K) and diverges from PC1. The correspondence that exists between Ca and lake level during high lake level is not anymore valid when erosional potential is high. This is why the Ca record cannot be used as lake level proxy.

#### Paleohydrological reconstructions since 1879

In the following section, we apply our age model to the entire sedimentary section of core AZA13-1.

The PC1 proxy indicates high runoff activity between 1880 and 1895 (Fig. 6). The T1M present in Facies 1 are composed of thick layers with a large amount of material derived from shorelines close to the forest (Fig. 4A). During this period, the diatom assemblage indicates increased freshwater inputs and a high lake level stand (Supplementary Figure 5). After 1900, the Lake Azigza sedimentary sequence is dominated by the deposition of T2M (Fig. 4B), suggesting lower lake levels. From 1900 to the 1950s, the runoff activity is low (Fig. 6). During the first half of the 20<sup>th</sup> century, the diatom assemblage is dominated by benthic species of Cyclotella, which suggests a lower lake level context. At the beginning of the 1960s, the runoff activity becomes higher and remains stable until the beginning of the 1980s. Several T1M are developed during the 1960s (Fig. 6), which argues in favour of a higher lake level. During the 1980s and the 1990s, the runoff activity is low. The absence of microstructure indicates that the runoff intensity is also substantially reduced. After the mid-1990s, the runoff activity increases gradually (Fig. 6). In the 2000s, and for the first time in the entire sequence, divergent trends are observed between the runoff activity and carbonates (Fig. 3 and Supplementary Figure 4B), which suggests a shift in the hydrosedimentary dynamics. Higher detrital input to the deep basin leads to a dilution of carbonates and organic matter in the sediment. From the beginning of the 1980s, we can also observe rapid fluctuations in the runoff activity proxy. These rapid bursts in runoff activity could reflect an increase in the inter-annual hydrological variability and/or in the erosional potential since the 1980s, both participating to increase the sensitivity of the runoff proxy. The

occurrence of T3M between 2008 and 2010 also highlights recent and rapid shifts in the hydro-sedimentary dynamics linked to significant hydrological changes in only two years (Fig. 6A). This result suggests that, when compared to the last 134 years, the major dry periods occurred since the beginning of the years 2000.

In the present study, we compare the lake sediment records with re-analysis of hydro-climate data (CRU) for the period 1901-2013 (Fig. 6). We also use the updated Palmer Drought Severity Index (PDSI) covering a longer period (Esper et al., 2007; Wassenburg et al. 2013). For the period 1879 to 1901, the PDSI reconstruction shows negative values indicating dry conditions (Fig. 6). This apparent discrepancy with our sedimentary proxy results could be due to uncertainties in the age model at the base of the sequence of core AZA-13-1 subjected to compaction effect due to coring processes. When using the CFCS model for the entire core, the age at the bottom is 1899 instead of 1879. However, close similarities in the K-XRF records from both cores AZA-13-3 and AZA-13-1 rather reinforces the age of 1879 at the base of sequence. It is also likely that the PDSI, a compilation of tree-ring width data from the Rif as well as the Middle and the High Atlas, reflects a regional hydro-climate signal. After 1900, PDSI and CRU data show approximately similar trends (Fig. 6). Negative PDSI values between the 1900s and the 1950s suggest dry conditions with some fluctuations, which is in agreement with our interpretations. At the beginning of the 1960s, the lake level rises by about 10 m in comparison with levels for the mid-1950s (Figs. 6 and 8). Precipitation and PDSI reach their highest values of the last 134 years, indicating much wetter conditions leading to high runoff activity.

Runoff activity decreases at the beginning of the 1980s, when lake level, PDSI and precipitation are minimal (Fig. 6). In the mid-2000s, the hydro-sedimentary dynamics of the lake reaches a tipping point as discussed in the previous section. This shift is supported by a

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strong increase of the PC1 (and K) for the period 2000-2013 (Fig. 7). The dry period from the 1990s onwards seems to end with an exceptionally low lake level in 2008. The occurrence of several dry episodes at the end of the 20<sup>th</sup> century have been also inferred from PDSI reconstructions (Esper et al., 2007). Although higher precipitation led to higher lake levels between 2009 and 2012, the level has dropped by about 3 m since 2013 (Figs. 6 and 8; Vidal et al., 2016). Consequently, the lake area appears to have been affected by several dry periods since the middle of the 1990s. These drier conditions could be the precursor signs of the impact of ongoing climate change on the precipitation regime in regions around the southern periphery of the Mediterranean, as expected for the 21<sup>st</sup> century (IPCC, AR5, 2013).

Earlier studies on deep-water sediment cores from Lake Azigza (Flower et al., 1989) failed to yield satisfactory paleohydrological reconstructions mainly due to the low resolution of the geophysical and chemical dataset and the macro-scale approach of the sedimentological observations. Other studies on Middle-Atlas lake sediments (Ifrah, Iffer and Afourgagh) (Damnati et al., 2012) suggest that wetter conditions prevailed in the early 20<sup>th</sup> century, followed by unstable conditions between 1920 and 1965. However, after 1965, it is difficult to assess the impact of climatic versus human factors on past lake levels and on the hydrogeochemistry.

The methodological approach used here is applied to the sedimentary record of a lake with minor human disturbance, calibrated against past lake-level changes and regional precipitation data. By considering changes in the sedimentary facies together with lake-level fluctuations, we present evidence for variations in the hydro-sedimentary dynamics of Lake Azigza over the last 134 years. This study improves our understanding of a lake sediment record mainly controlled by detrital inputs and autochthonous carbonates with limited post-depositional processes, reflecting past hydrological variability at inter-annual to decadal time-

scales. As such, our study contributes valuable insights for the interpretation of lake infill sequences and can be applied to a wide range of lacustrine systems which exhibit substantial water-level fluctuations.

### Conclusions

We show that the Lake Azigza sedimentary record is useful for studying the hydrological variability of the Middle-Atlas region. Based on microfacies and XRF elemental analyses of the sediments, we provide proxies of runoff activity and lake-level changes. The onset and duration of the response to drier and wetter periods at the Lake Azigza site is well represented by detrital element contents (K, Si, Ti and Fe) that are considered as proxies of runoff activity. The sedimentary facies have specific geochemical signatures attributed to hydro-sedimentary processes (related to variable runoff activity) during high and low lake level periods. These lake-level changes are inferred from sediment microstructures linked to the runoff intensity and distance from the shoreline.

During high-precipitation periods, the surface runoff is increased, associated with a rise in the lake water level, implying that more detrital minerals and runoff-induced autochthonous carbonates are transported to the deep basin of the lake.

The sedimentological proxies are calibrated with regional hydro-climate data for the period 1963-2013 and then interpreted for the last 134 years. The sediment record faithfully reproduces the hydrological fluctuations observed since 1879 at decadal scales. We show that the sensitivity of the PC1 index is increased after periods of drought (e.g. low lake level). The strong response of the runoff proxy for the last decade suggest that the major low lake level reached in 2008 could be the first sign of increased drought duration expected for the period 2021-2050 in the Middle-Atlas mountains. Moreover, calibration of dated sediment sequences

with local precipitation data is crucial for understanding the complex hydro-sedimentary dynamics of lacustrine systems and developing reliable proxies of hydrological variability.

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### **Figure captions**

**Table 1:** A) Correlation coefficients (R) between XRF elements used for the PCA for the entire AZA-13-1 sequence. B) R of all elements in PC1 and PC2.

**Figure 1**: A) Geographical position of Lake Azigza in the Moroccan Middle Atlas (black circle). B) Photograph of the lake in 2013 (Google Earth). C) Shorelines at the southeastern end of the lake. White line indicates highest visible limit between subaquatic and subaerial sediments next to the shorelines and white arrows highlight the shorelines inclination during low and high lake levels respectively). D) Gullies on the southern lake shore (white arrows). E) Photographs of cores AZA-13-1, AZA-13-3 and AZA-13-4 showing core length (cm). F) High-resolution bathymetry of the lake (derived from kriging method by interpolation on 150,000 points obtained with the Humminbird single beam sonar). Position of cores is indicated (black circle).

Figure 2: Age-depth model for cores AZA-13-3 and AZA-13-1.

A) <sup>226</sup>Ra, <sup>210</sup>Pb, and <sup>137</sup>Cs profiles in core AZA-13-3. The position of the identified peak of Nuclear Weapon Testing (NWT) corresponding to 1963 AD is indicated on the <sup>137</sup>Cs profile. Sedimentation rates (CFCS) obtained from the <sup>210</sup>Pb profile and the <sup>137</sup>Cs peak. B) Stratigraphic correlations between cores AZA-13-3 and AZA-13-1 based on K-XRF records. C) Age-model of core AZA-13-1 derived from a CFCS model between 0 and 28 cm (2013-1963) using a mean sedimentation rate of 0.56 cm.y<sup>-1</sup>, and linear interpolations between correlations for the interval 28 cm - 63.5 cm (1963-1879).

Figure 3: Sedimentological and geochemical observations of core AZA-13-1.

From left to right: down-core distribution of facies; natural-light images of thin-sections; core photograph; radiography; distribution of type 1, 2 and 3 microstructures (asterisks); TOC (%) (Total Organic Matter) in bulk sediment and inc/coh ratio; CaCO<sub>3</sub>% content in bulk sediment and Ca-XRF intensity; PC2; Sr/Ti - XRF ratio; % silt fraction in sediment (black symbols indicate co-variation with TOC% at the base of the sequence, red symbols indicate co-variation with CaCO<sub>3</sub>%); % clay fraction in sediment, Fe-XRF, Ti-XRF, K-XRF, PC1 and Si-XRF.

Figure 4: Microscale observations of core AZA-13-1

Left: Natural light thin-sections of core AZA-13-1 with down-core distribution of microstructures: type 1 (red rectangles), 2 (black rectangles) and 3 (grey rectangles). A) type-1 microstructures: cross-polarized light (XP) images showing discontinuity in sedimentary sequence. Coarser materials are visible in natural light (NL) image of the microstructure (a). High amounts of wood fragments (c) and ostracods (back-scattered electron - BSE - image in b) are present. EDS elemental mapping on indurated blocks (right hand side) shows calcitic shells of ostracods. The mapping is performed for Ca, Si and Fe in each case. B) type-2 microstructures: NL image of thin section showing ostracod-rich layers. Note the absence of wood fragments in these structures and the large size of the calcitic bivalve shells. C) type-3 microstructures: XP image of thin section in the laminations at the top of the sequence. BSE image and EDS analyses (a) of laminations showing alternation of carbonate-rich and detrital layers containing angular quartz grains (b).

**Figure 5**: From left to right: photographs of core and indurated blocks, with SEM-EDS mapping of a and c: coarse detrital facies in middle of the sequence: d and b: sand-sized detrital grains and calcitic shells of ostracods and other bivalves.

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Figure 6: Runoff activity and lake water level changes versus precipitation and drought severity index.

From the bottom to top: down-core distribution of facies. C) Position of lake-level indicators: type 1, 2 and 3 microstructures (asterisks; see legend in Fig. 3) and diatom valve concentrations compared to 2013. B) Ca and PC1 values (units expressed in factor scores) derived from AZA-13-1 age model. Assuming a mean sedimentation rate of about 0.56 cm.year<sup>-1</sup>, the moving average of six measurements records the inter-annual variability. Black arrows highlight opposite trend for PC1 and Ca. A) Annual precipitation CRU-data (deviation from the mean calculated for the whole period 1901-2013). Lake level relative to the reference level of 2013 (red squares) derived from photographs available in Gayral and Panouse (1954), and lake-level measurements in Flower et al. (1989) and Flower and Foster (1992). Palmer Drought Severity Index (PDSI) from Esper et al. (2007) and Wassenburg et al. (2013). Negative values reflect drier conditions and vice versa.

**Figure 7:** Annual K-XRF and PC1 versus precipitation for the period 1963-1999 (black and grey symbols and associated regression line) and for the period 2000-2013 (orange and green).

**Figure 8**: Aerial photographs of the lake in (A) 1964 and (B) 1973 obtained from the ANCFCC (National Agency of Land Conservation, Cadastre and Cartography, Rabat, Morocco). White lines correspond to the delimitation of shorelines in 2013. (C) Photograph of the lake in 2008 (view from the western side of the lake). The low lake level is marked by dewatering of the western shallow basin. (D) Google earth satellite map of the lake in 2016.

### Supplementary Figure captions

Supplementary Table 1: Hydrogen Index (HI) and Oxygen Index (OI) values from Rock Eval analysis in core AZA-13-3.

Supplementary Figure 1: Laser diffraction grain size results in core AZA-13-1. Diameters are expressed in micrometers and plotted on a logarithmic scale.

Supplementary Figure 2: X-ray diffraction spectrum of total mineralogy of sediments in core AZA-13-1.

Supplementary Figure 3: X-ray diffraction spectrum of clay mineralogy of sediments in core AZA-13-1.

Supplementary Figure 4: A) PCA analyses of XRF data for the entire AZA-13-1 sequence, with the identification of three chemical end-members autochthonous carbonates (red oval), detritism (blue oval) and organic matter (black oval); B) PC1 versus PC2 values (white, black and brown dots correspond to sedimentary Facies 1, 2 and 3 respectively). Correlation coefficients for positive and negative relationship between PC1 and PC2 are indicated in the figure.

Supplementary Figure 5: Details of the methodology and results of the diatom analysis in core AZA-13-1.