



# Palaeomagnetism for chronologies of recent alpine lake sediments: successes and limits

C. Crouzet · B. Wilhelm · P. Sabatier · F. Demory · N. Thouveny ·  
C. Pignol · J.-L. Reyss · O. Magand · A. Jeltsch-Thömmes · M. Bajard ·  
L. Augustin · F. Arnaud

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**Abstract** Chronologies of lake-sediment records covering the last centuries to millennia are usually based on both short-lived radionuclides and radiocarbon dating. However, beyond the range of short-lived radionuclides, age model accuracy often suffers from large radiocarbon uncertainties. For high-altitude

records, this issue is even more prominent as terrestrial plant fragments for radiocarbon dating are often lacking due to the sparse vegetation in such environments. In this study, we evaluate the potential of the geomagnetic field secular variations as a complementary tool to establish more robust age–depth relationships. Our palaeomagnetic study, applied to five high-altitude lakes from the western European Alps, first shows that recent unconsolidated sediments can carry stable remanent magnetization. The analysis of the magnetic parameters indicates that low-coercivity

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C. Crouzet (✉)  
Université Savoie Mont Blanc, Université Grenoble Alpes, CNRS, IRD, IFFSTAR, ISTERre,  
73376 Le Bourget Du Lac, France  
e-mail: christian.crouzet@univ-smb.fr

B. Wilhelm · O. Magand  
Université Grenoble Alpes, CNRS, IRD, Grenoble INP (Institute of Engineering Univ. Grenoble Alpes), IGE,  
38000 Grenoble, France

P. Sabatier · C. Pignol · M. Bajard · F. Arnaud  
Université Grenoble Alpes, Université Savoie Mont Blanc, CNRS, UMR 5204, EDYTEM,  
73000 Chambéry Cedex, France

F. Demory · N. Thouveny  
Aix Marseille Univ, CNRS, IRD, INRA, Coll France, CEREGE, Aix-En-Provence, France

J.-L. Reyss  
LSCE, Université de Versailles Saint-Quentin CEA-CNRS, avenue de la terrasse,  
91198 Gif-Sur-Yvette Cedex, France

A. Jeltsch-Thömmes  
Climate and Environmental Physics, Physics Institute, University of Bern, Bern, Switzerland

A. Jeltsch-Thömmes  
Oeschger Centre for Climate Change Research, University of Bern, Bern, Switzerland

M. Bajard  
Centre for Earth Evolution and Dynamics, University of Oslo, PO Box 1028, Blindern, 0315 Oslo, Norway

L. Augustin  
Division technique de l'INSU, Centre de Carottage et de Forage National, CNRS, La Seyne sur Mer, France

pseudo-single domain magnetite grains carry the natural magnetization. Nevertheless, the quality of palaeomagnetic secular variation records varies from one lake to another. This quality can be illustrated through the calculation of the declination/inclination maximum angular variations and their comparison to the expected value. Compared with available models, the declination variations are usually too large and the inclination too high. We discuss the validity of palaeosecular variation (PSV) of the Earth's magnetic field regarding rock magnetism, magnetization processes and possible deformation during coring. From a magnetic point of view, the quality of data is variable, but the characteristic remanent magnetization direction is consistent at site level between neighbouring lakes and with the reference curve, suggesting that geomagnetic field secular variations are approximately recorded. Finally, we attempt to correlate the declination/inclination variations of the characteristic remanent magnetization measured in the five records to the reference geomagnetic model to provide additional chronological markers for age–depth modelling. These stratigraphic chrono-markers appear in systematic agreement with our previous chronological data and enable a reduction of dating uncertainties up to 30% when including these chrono-markers in the age–depth modelling. This agreement supports the interpretation that PSV may have been recorded more or less accurately depending on the studied lake. Therefore, coupled with a comprehensive understanding through other analysis (sedimentology, dating, geochemistry), PSV can be used to improve the age models in the more favourable cases.

**Keywords** Paleomagnetism · Secular variation · Lake sediment · Dating · Magnetization · Alps

## Introduction

A still-challenging point for palaeoenvironmental or palaeoclimatic studies carried out using lake sediment archives is the accuracy of chronologies. For the last century, short-lived radionuclides ( $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$ ) allow reliable dating (Appleby 2008). Beyond this period, chronologies are mostly based on varve counting or more often on  $^{14}\text{C}$  dating of terrestrial plant fragments. However, such remains may be scarce or absent in

high-altitude and high-latitude lakes or during pleni-glacial periods. Moreover,  $^{14}\text{C}$  dating often implies relatively large uncertainties over the last centuries because of age plateaus (Sabatier et al. 2010) caused by a strong increase in atmospheric  $^{14}\text{C}$  production (Reimer et al. 2013). Therefore, developing another independent chronological tool appears essential to improve the chronologies of lake-sediment records (Wilhelm et al. 2018).

Since the pioneering works of the 1970s (Creer et al. 1972; Thompson and Kelts 1974) and the establishment of the first palaeomagnetic secular variations (PSV) curve from UK lakes (Thompson and Turner 1979), the development of limno-magnetism has rapidly grown. The main topic of most studies is the construction of PSV over a certain time span to better constrain geomagnetic field models (Lougheed et al. 2014). In the last decade, the development of global models for Holocene geomagnetic field variations (ARCH3k, Korte et al. 2009 or CALS10k, Korte et al. 2011) has made it possible to compare geomagnetic model outputs with PSV recorded worldwide in marine or lacustrine sediments. In particular, the occurrence of several changes in the trend of the PSV during the Holocene allows test matching of the reference curve with the directions of the measured natural remanent magnetization (NRM) and then the establishment of depth/age couples. Recently, some studies have successfully applied this approach to improve chronologies of records from the Arctic (Barletta et al. 2010), China (Yang et al. 2013), North America (Lund and Platzman 2015), on Italian crater lakes and marine cores (Rolph et al. 2004; Vigliotti 2006) and the western French Alps (Bajard et al. 2016; Wilhelm et al. 2016a).

PSV records in lake sediments are considered reliable when the following conditions are fulfilled: (1) a stable magnetic mineralogy; (2) the demonstration that the remanent magnetization is of depositional origin (i.e., orientation of magnetic grains in the soft sediment); and (3) the good record and preservation of the direction of magnetization (i.e., no destruction of sedimentary structures). Fulfilling these conditions implies that the paleomagnetic record represents a primary magnetization. Demonstrating this point is challenging and efforts remain to better understand how sediments become magnetized.

PSV reconstruction based on lake sediments suffers from many problems linked to the magnetic

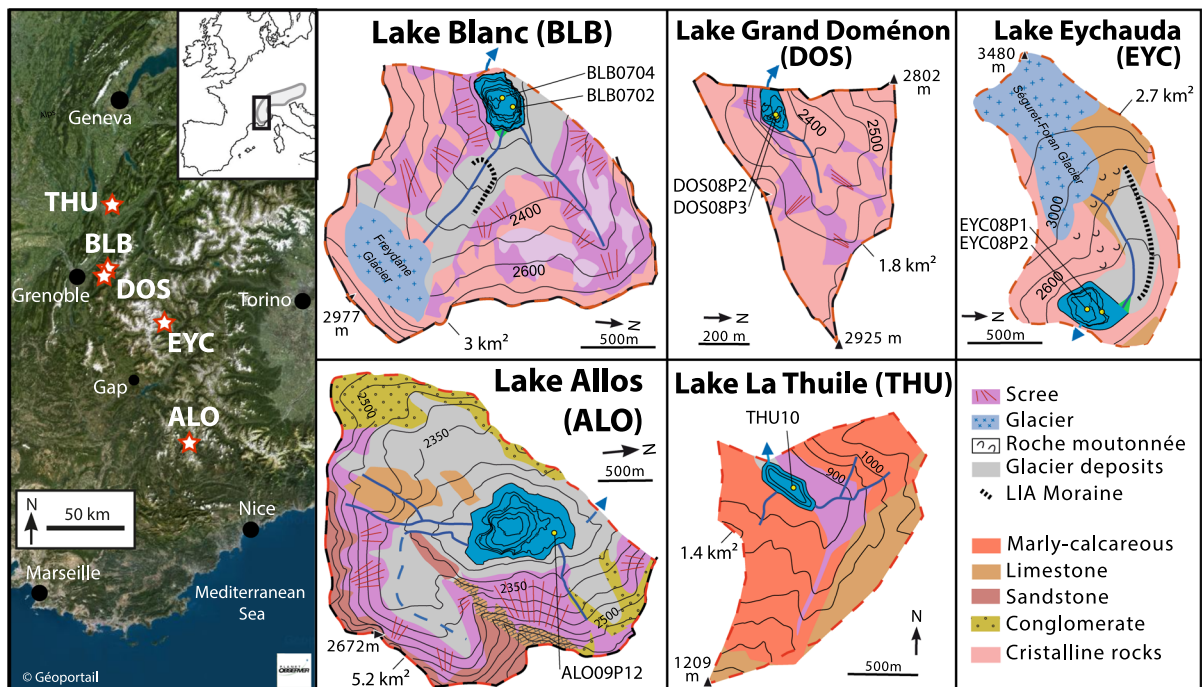
mineralogy and its ability to record the Earth's magnetic field. In addition, post-depositional processes, such as (bio)chemical magnetization acquisition, may affect the quality of the PSV record (Roberts et al. 2012). Finally, instantaneous deposits, such as those from flash floods and gravitational processes or seismites as well as coring and sampling deformations (Ali et al. 1999), are also responsible for directional mistakes or poor recording/preservation of the magnetization. To solve these problems, careful magnetic mineralogy must be established, but it is not sufficient. The process of post-depositional remanent magnetization acquisition and, by extension, lock-in depth must be understood (Spassov and Valet 2012). Additionally, cleaning of the PSV record based on rejecting questionable levels can be attempted (Demory et al. 2005).

Thanks to several sedimentological studies and the availability of well-dated cores, this study presents five lake-sediment records from high-altitude catchments of the western European Alps. The objective is to explore the possibility to improve the chronologies of few alpine lakes by using the paleomagnetic data.

## Study sites

Five mountain lakes located in small high-elevation (880 m to 2517 m a.s.l.) catchments (1 to 6 km<sup>2</sup>) of the western European Alps are studied (Fig. 1): La Thuile (880 m a.s.l., 45°31'50.63"N, 6°3'39.9"E, Bajard et al. 2016), Allos (2230 m a.s.l., 44°14'N, 6°42'35"E, Wilhelm et al. 2012a), Blanc Belledonne (2170 m a.s.l., 45°10'42"N, 5°58'21"E, Wilhelm et al. 2012b), Doménon (2390 m a.s.l., 45°9'46.81"N, 5°57'45.20"E, Wilhelm et al. 2016b), and Eychauda (2517 m a.s.l., 44°55'55.61"N, 6°28'49.02"E, Wilhelm 2012).

The catchments of Lakes La Thuile and Allos are dominated by sedimentary rocks (limestone and sandstone), while those of Lakes Blanc Belledonne, Doménon, and Eychauda are composed of crystalline rocks (amphibolite and gneiss). The catchment of Lake La Thuile is covered partly by a dense forest and partly by cultivated fields. The catchment is poorly drained, and the two small inflows are identifiable only during heavy precipitation. The sediments of Lake La Thuile are silty-clayey organic-rich mud finely laminated in the lower part of the sequence, in which some



**Fig. 1** Location of the studied sites in the French Alps on the left. Hydrological, geomorphological and geological characteristics of the studied lakes and respective catchments on the right

thick event layers are identified (Bajard et al. 2016). Due to the high elevation, the vegetation around the catchments of Lakes Allos, Blanc Belledonne, Doménon and Eychauda is made of sparse grasslands. This condition may explain the lack of plant fragments for  $^{14}\text{C}$  dating. In addition, this elevation made possible the development of glaciers, which resulted in large amounts of easily erodible glacial deposits in the Allos, Blanc Belledonne and Eychauda catchments. The consequence is a high detrital input for these lakes, as suggested by well-developed deltas (Wilhelm 2012). For these three lakes, the sediment is indeed composed mainly of fine-grained (clays and fine silts) detrital material. In addition, event layers are very frequent in these sequences and result from floods or subaquatic landslides. By contrast, there is no evidence of glacial deposits in the catchment of lake Doménon, and only a small and grass-covered delta is present for this lake. As a result, a lower detrital input is observed. The sediment appears as a mix of detrital and organic materials (values of total organic carbon reaching 10%), and no clear event layers are observed (Wilhelm 2012).

## Materials and methods

### Coring and sediments analyses

In Lakes Allos (ALO), Blanc Belledonne (BLB), Doménon (DOS), and Eychauda (EYC), a UWITEC gravity corer was used to retrieve short sediment cores from the deepest part of the lake basins. Information about coring campaigns can be found in the French Cyber-Core Repository at [www.cybercarotheque.fr](http://www.cybercarotheque.fr). For palaeomagnetic analysis, several cores were used, including the following: ALO09P11 (length = 80 cm, IGSN: IEFRA000H), ALO09P12 (length = 140 cm, IGSN: IEFRA000I), BLB0702 (length = 40 cm, IGSN: IEFRA05JK), BLB0801 (length = 103 cm, IGSN: EDY000003), BLB1204 (length = 113 cm, IGSN: IEFRA003I), DOS08P2 (length = 63 cm, IGSN: IEFRA006T), DOS08P3 (length = 83 cm, IGSN: IEFRA006U) and EYC08P2 (length = 106 cm, IGSN: IEFRA006 W). For Lake La Thuile, a long core of 19 m was retrieved using a piston corer.

The studied part of the core THU10 (length = 500 cm, IGSN: IEFRA00BB) is composed of 5 sections, plus a short core taken to provide a well-preserved

sample of the water–sediment interface. We constructed the composite sequence using the distinctive beds that appear in each section to ensure a precise and reliable stratigraphic correlation (Bajard et al. 2016). All cores were split lengthwise and photographed for high-resolution ( $20 \text{ pixels mm}^{-1}$ ) visual analysis of sedimentary facies. In all the studied lakes, the occurrence of well-preserved laminations in the sediments suggests the absence of bioturbation.

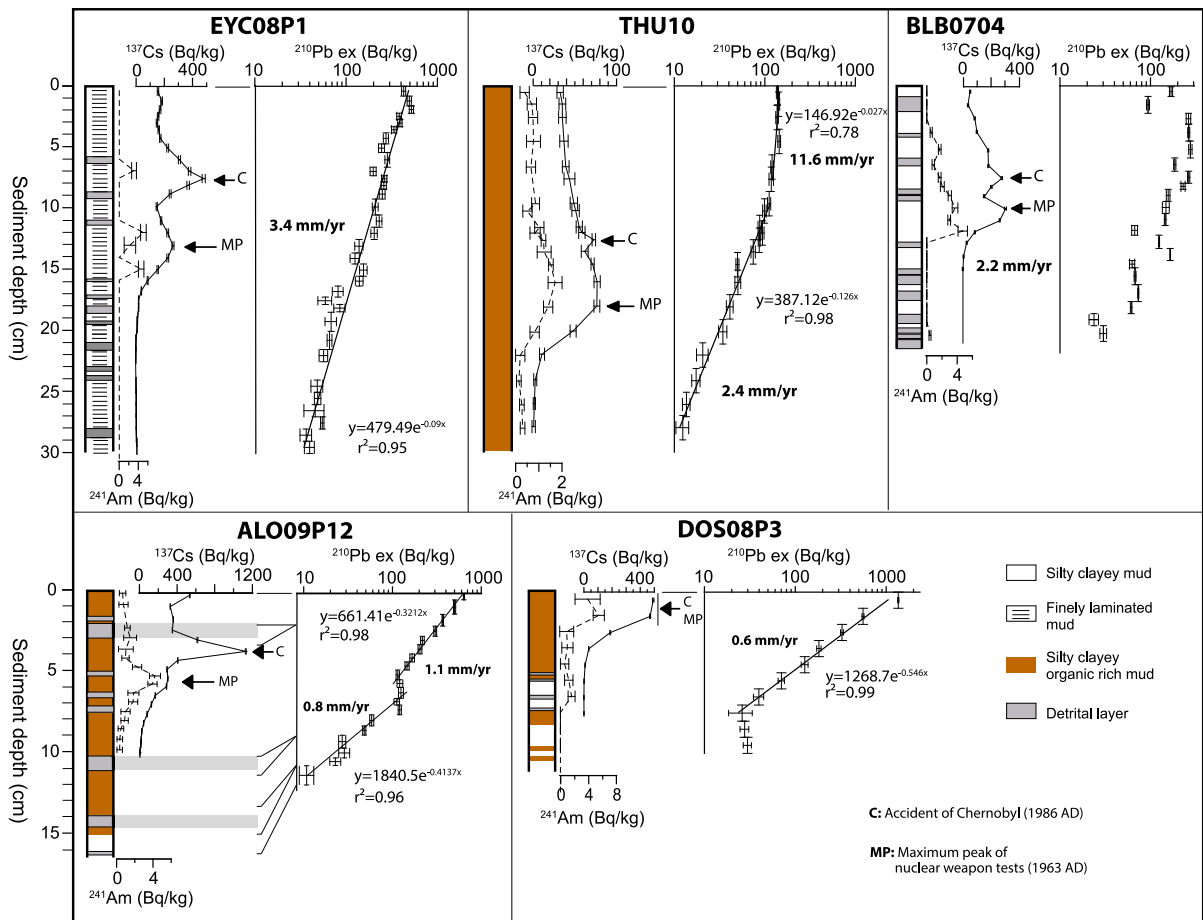
### Dating methods

#### *Short-lived radionuclides*

We measured short-lived radionuclides ( $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$  and  $^{241}\text{Am}$ ) in the uppermost part of cores for all sequences, following a continuous irregular sampling step of 0.5 to 1 cm adapted to lithological variations (Fig. 2). Measurements were made by gamma spectrometry using high-efficiency, very low background, well-type Ge detectors in the Modane underground laboratory (Reyss et al. 1995) for sediments from Lakes Allos, Doménon and La Thuile and in the IGE laboratory at Grenoble for Eychauda and Blanc Belledonne sediments (Wilhelm et al. 2012b). In each sample of cores, the  $^{210}\text{Pb}$  activity was calculated by subtracting the  $^{226}\text{Ra}$  activity (determined as  $^{210}\text{Pb}$ -supported) from the total  $^{210}\text{Pb}$  activity. We then used the constant flux/constant sedimentation (CFCS) model to calculate the sedimentation rate (Goldberg 1963). Furthermore, two peaks are expected from the  $^{137}\text{Cs}$  activity profiles, corresponding to the Chernobyl accident in AD 1986 and to the maximum nuclear fallout in AD 1963, while one peak of  $^{241}\text{Am}$  activity is expected for the atmospheric nuclear weapons tests (Appleby et al. 1991).

#### *Historical lead (Pb) contaminations*

Historical lead (Pb) contaminations were used as chronological markers for the Lake Blanc Belledonne sedimentary sequence (Wilhelm et al. 2012b). Samples from core BLB0704 (1-cm sampling steps) were analysed for trace elements using conventional XRF methods at the IFREMER marine geology laboratory of Brest. To disentangle natural and human-induced changes in Pb concentration, they were normalized to yttrium (Y), a lithophile element with comparable



**Fig. 2**  $^{137}\text{Cs}$  activity and  $^{210}\text{Pb}$  activity profiles for cores EYC08P1, THU10, BLB0704, ALO09P12 and DOS08P3, with estimation of MAR based on the constant flux/constant sedimentation (CFCS) model. The  $^{210}\text{Pb}$  activity profile of core BLB0704 is disturbed by the numerous flood events,

making the application of CFCS model unsuitable (Wilhelm et al. 2012b). Note that the CFCS model was applied to core ALO09P12 on the synthetic sediment depth (without the thick graded beds considered as instantaneous deposits) (Wilhelm et al. 2012a)

concentration and geochemical behaviour. Detailed results are presented in Wilhelm et al. (2012b).

#### Correlation to historical events

Historical earthquakes and extreme floods can result in particular deposits, here called event layers. Linking such layers with their respective triggering historical events may thus yield additional chronological markers (Monecke et al. 2004). This approach was undertaken for the Lake Blanc Belledonne sedimentary sequence, allowing linkage of 10 historical floods to 10 event layers and thereby adding 10 chronological markers over the last 270 years (Wilhelm et al. 2012b, 2016b; ESM1).

#### $^{14}\text{C}$ dating

Due to the high elevation, the vegetation of the catchments of Lakes Allos, Blanc Belledonne, Doménon and Eychauda consists of sparse grasslands. This situation may explain the difficulty finding plant fragments for  $^{14}\text{C}$  dating. Small-size plant macroremains were sampled for Lakes Doménon, Allos, Blanc Belledonne and La Thuile. Identifiable pine needles, buds, twigs and leaves were separated and used for accelerator mass spectrometry at the Poznan Radiocarbon Laboratory and at the Laboratoire de Mesure  $^{14}\text{C}$  (LMC14) ARTEMIS at the CEA (Atomic Energy Commission) Institute at Saclay.  $^{14}\text{C}$  ages were calibrated (Intcal13 calibration curve, Reimer et al.

2013). No vegetal macroremains were found in Lake Eychauda sediments and only one in Lake Blanc Belledonne sediments, certainly due to the very sparse or absent vegetation in their high-altitude catchments.

#### *Age–depth models*

Before proceeding to the age–depth modelling, a composite depth called “event-layer free depth” was built by removing these layers because they were interpreted as instantaneous deposits. Age–depth models were built using all the above dating methods. The models were generated using a smooth spline function included in the R-code package “clam” (Blaauw 2010).

#### *Magnetic measurements and analyses*

All the magnetic measurements were performed in the CEREGE laboratory (Aix-Marseille University). The low field magnetic susceptibility was measured routinely using a Bartington MS2 contact sensor on split cores at a 5-mm resolution. In each sedimentary unit, cubic samples were collected to perform anisotropy of magnetic susceptibility measurements. The latter are used to ensure the good preservation of sedimentary features. The measurements were performed using the AGICO MFK1-FA Kappabridge (spinning specimen method).

U-channels subsamples were taken from the cores to perform measurements of remanent magnetizations. The magnetizations were measured using a superconducting quantum interference device (SQUID) pass-through cryogenic magnetometer (2G 760R) located in a shielded room. Each point of measurement integrates a signal of 4-cm width along the X and Y axes, and 7 cm along Z axis. U-channel samples were subjected to stepwise alternating field (AF) demagnetization of the natural remanent magnetization (NRM) using steps of 10 mT until the measured remanence was less than 20% of the NRM, usually between 60 and 80 mT.

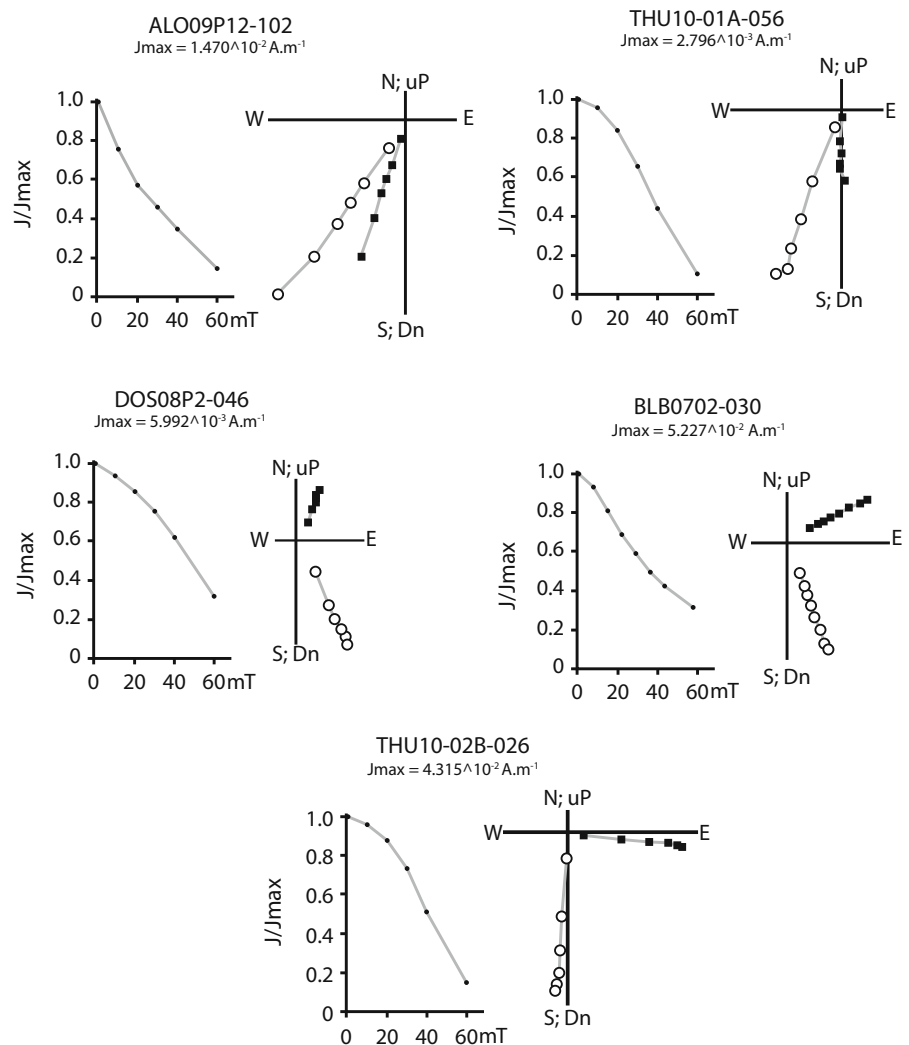
The NRM was carefully analysed using demagnetization curves, orthogonal diagrams and consistency of successive individual measures (Fig. 3). Principal component analyses have been performed using the PuffinPlot software (Lurcock and Wilson 2012) to calculate the Characteristic Remanent Magnetization (ChRM). A careful examination of demagnetization

diagrams shows a unidirectional behaviour indicated by linear decay to the origin over a range of AF treatments. The mean angular deviation is usually lower than 6, revealing good stability of the magnetization direction. When the mean angular deviation is higher than 6, the measurement point is eliminated.

To identify magnetic particles, different measurements were performed in addition to grain-size distribution. To distinguish different mineralogical and grain-size fractions within the magnetic components, two types of laboratory remanent magnetizations were artificially imparted: isothermal remanent magnetization (IRM) and anhysteretic remanent magnetization (ARM). ARM was produced in-line along the U-channel axis, using a 100-mT alternating field with a superimposed 0.05-mT steady field. For ARM, demagnetization was done following steps of 10, 20, 30 and 40 mT. IRM was obtained by passing the U-channels through two different Halbach cylinders that develop fields of 1 T and 0.3 T. Additionally, environmental magnetic parameters such as the S-ratio ( $= 0.5 \cdot (1 - (IRM_{-0.3T}/SIRM_{1T}))$ ) from Bloemen-dal et al. (1992), the ratio ARM/saturated IRM, and the ratio ARM/low field magnetic susceptibility were calculated. AF demagnetization data were used to compute the median destructive fields of the NRM, ARM and IRM. All remanence measurements (NRM, ARM and IRM) realized on U-channels were performed at intervals of 2 cm.

Additionally, several classic magnetic experiments were conducted on selected samples. A small quantity of sediment was collected in each characteristic stratigraphic layers to study the hysteresis properties using a MicroMag Vibrating Sample Magnetometer (VSM—Princeton Measurement Corporation) and to perform the Lowrie-Fuller test. The susceptibility versus temperature was monitored using a CS3 furnace combined with KLY3 s apparatus from AGICO. Sixteen samples were chosen for 3-axis IRM acquisition using 0.1, 0.3 and 3 T fields and subsequent thermal demagnetization.

**Fig. 3** Alternating Field (AF) demagnetization of Natural Remanent Magnetization (NRM) represented as Orthogonal diagrams for characteristic samples showing only one component



## Results

### Chronology

#### Short-lived radionuclides ( $^{210}\text{Pb}$ , $^{137}\text{Cs}$ and $^{241}\text{Am}$ )

( $^{210}\text{Pbex}$ ) profiles for cores EYC08P1 and DOS08P3 showed constant decreases to very low values ( $< 20 \text{ Bq kg}^{-1}$ ). For these two lakes, the CFCS model gave sedimentation rates of  $3.4 \pm 0.3$  and  $0.57 \pm 0.02 \text{ mm yr}^{-1}$  for EYC08P1 and DOS08P3, respectively (Fig. 2). The  $^{137}\text{Cs}$  and  $^{241}\text{Am}$  activities peaks in core EYC08P1 at 7.5 and 13 cm can be attributed to the Chernobyl fallout in AD 1986 and the maximum nuclear tests in AD 1963. These two chronological markers agree with the  $^{210}\text{Pbex}$ -derived

chronology. The  $^{137}\text{Cs}$  activity and  $^{241}\text{Am}$  activity profiles of cores DOS08P3 reveal a large peak in the top 2 cm. This peak may be explained by the low sedimentation rate and the low sampling step, which both make the two expected peaks impossible to distinguish.

The  $^{210}\text{Pbex}$  activity profile of core THU10 shows two distinct point alignments with a slight decrease for the top 8.5 cm, followed by a marked decrease from 8.5 to 29 cm (Fig. 2). Using the CFCS model on the upper and lower parts of the profile, the  $^{210}\text{Pbex}$  activity indicates sedimentation rate values of  $11.6 \pm 1.5$  and  $2.4 \pm 0.2 \text{ mm yr}^{-1}$ , respectively. Peaks of  $^{137}\text{Cs}$  activity related to the Chernobyl accident and of  $^{137}\text{Cs}$  activity and  $^{241}\text{Am}$  activity from the nuclear test fallout were found at 13 and 18 cm,

respectively, and are in good agreement with  $^{210}\text{Pb}_{\text{ex}}$  activity chronology (Bajard et al. 2016).

The  $^{210}\text{Pb}_{\text{ex}}$  activity profile for core ALO09P12 shows a regular decrease, punctuated by sharp dips to low values for three layers (2.1–3.3 cm, 10.2–11.3 cm and 13.2–14.9 cm, not shown) of thick, graded deposits (Wilhelm et al. 2012a). We exclude these  $^{210}\text{Pb}_{\text{ex}}$  activity values associated with flood deposits to construct a synthetic sedimentary record (Fig. 2). The resulting  $^{210}\text{Pb}_{\text{ex}}$  activity profile reveals two distinct point alignments. Using the CFCS model on each part of the profile indicates sedimentation rate values of  $1.1 \pm 0.1$  and  $0.8 \pm 0.1$  mm yr $^{-1}$  for the upper 8 cm and the lower section of the core, respectively. Peaks of  $^{137}\text{Cs}$  activity were found at 4 and  $5.5 \pm 0.5$  cm, with  $^{241}\text{Am}$  activity for the older peak, which confirms the Lake Allos age–depth model (Wilhelm et al. 2012a).

The  $^{210}\text{Pb}_{\text{ex}}$  activity profile for core BLB0704 presents a chaotic trend (Fig. 2), certainly associated with the frequent occurrence of event layers. This feature means that the CFCS model is not applicable to this sequence (Wilhelm et al. 2012b). Peaks of  $^{137}\text{Cs}$  activity are found at 7.5 and 10 cm, and  $^{241}\text{Am}$  activity is present at the depth of the deeper peak (Fig. 2) suggesting a sedimentation rate of 2.2 mm yr $^{-1}$ . These chronological markers agree with the peak of lead found at 8 cm (not shown) and linked to the maximum of contamination by leaded gasoline in AD

1974 (Wilhelm et al. 2012b). Finally, the association of 10 event layers with 10 historical floods that occurred in AD 1948, 1933, 1924, 1912, 1906, 1852, 1851, 1831, 1784 and 1748 allows building a first chronology over the last three centuries (Wilhelm et al. 2012b; ESM1). A stratigraphical correlation between cores BLB0704 and BLB1204 has been performed based on the noticeable event layers, allowing the reliable application of the chronological constraints of core BLB0704 to core BLB1204 (ESM1).

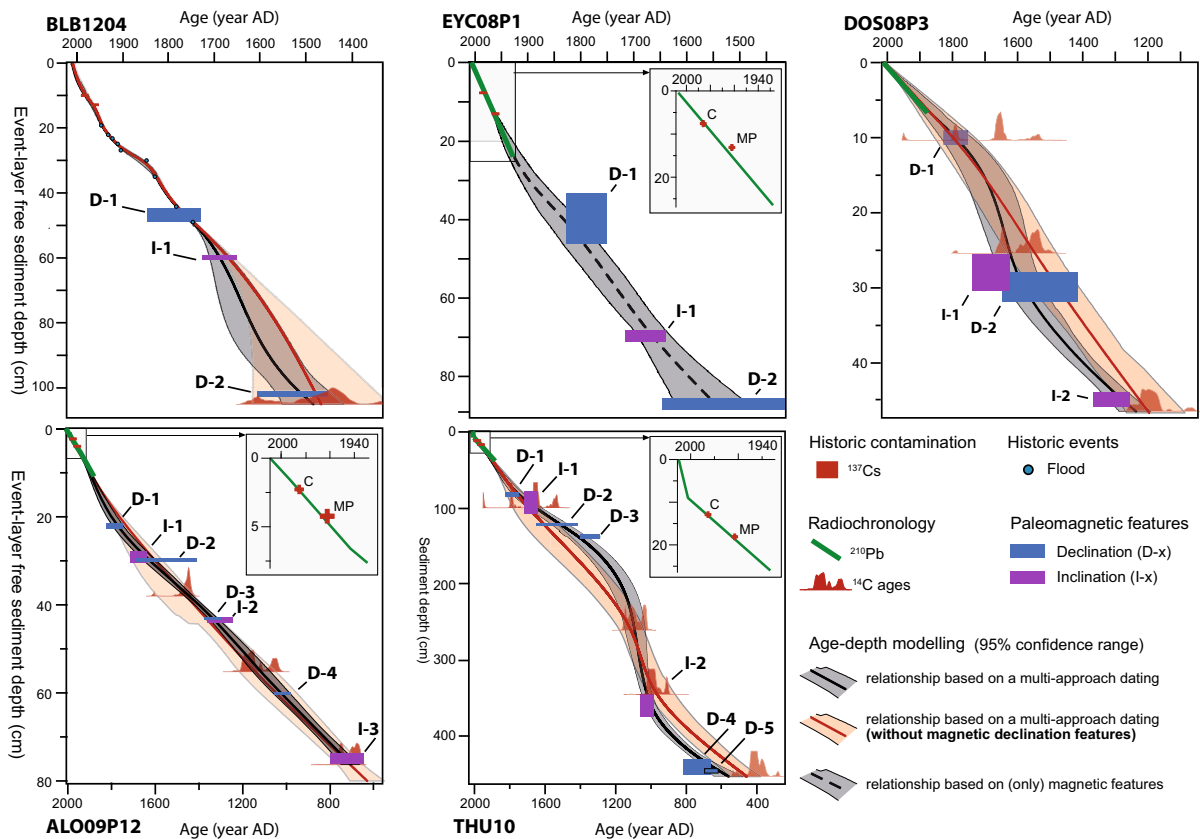
### First age–depth modelling

For sediment sequences THU10, DOS08P2, BLB1204 and ALO09P12, we combined the short-lived radionuclide results and the  $^{14}\text{C}$  ages (Table 1) to construct an age–depth model. For cores ALO09P12 and BLB1204, the flood layers were removed to construct a synthetic sedimentary record (Arnaud et al. 2002; Wilhelm et al. 2012a and b). Age–depth relationships calculated for each sequence revealed that ALO09P12 covers the last  $1400 \pm 75$  years cal BP; DOS08P2, the last  $1200 \pm 200$  years cal BP; THU10, the last  $1600 \pm 80$  years cal BP; and BLB1204, the last  $500 \pm 70$  years cal BP (Fig. 4). Hence, the application of the classic dating methods (short-lived radionuclides, correlation to historic events and  $^{14}\text{C}$  dating) delivered relatively large dating uncertainties

**Table 1** Radiocarbon dates for lakes Blanc Belledonne, Doménon, Allos and La Thuile sequences

Lake	Lab. code	Core depth (cm)	Composite sedimentary depth (cm)	Radiocarbon age $^{14}\text{C}$ yr. BP	Calibrated yr. BP (2 $\sigma$ )	Calibrated yr. AD (2 $\sigma$ )
DOS	SacA 32332	10–11	–	$250 \pm 30$	0–428	1522–1950
DOS	SacA 32333	25–26	–	$285 \pm 30$	160–456	1494–1790
DOS	SacA 32334	46–47	–	$850 \pm 30$	690–897	1053–1260
ALO	SacA 21326	71–73	37.8	$430 \pm 30$	335–529	1421–1615
ALO	SacA 24170	151–152	55.4	$880 \pm 30$	730–907	1043–1220
ALO	SacA 21330	130–132	76.3	$1295 \pm 30$	1177–1287	663–773
THU	SacA 24161	98–102	100	$250 \pm 30$	0–428	1522–1950
THU	SacA 24162	75–81	260	$960 \pm 30$	795–929	1021–1155
THU	Poz 41624	67–68	345.5	$1050 \pm 30$	924–1052	898–1026
THU	SacA 24163	77–79	450	$1630 \pm 30$	1415–1602	348–535
BLB	Poz 58121	104–106	–	$450 \pm 70$	419–558	1392–1531

For the lakes Allos and Blanc Belledonne we calculated the composite depth by removing the graded beds, witches were considered to be instantaneous deposits (Wilhelm et al. 2012a). For the lake La Thuile the composite depth corresponds to the depth of the master core (see Bajard et al. 2016)



**Fig. 4** Age–depth models for cores EYC08P1, THU10, BLB1204, ALO09P12 and DOS08P3, combining available data for each sequence among the historic  $^{137}\text{Cs}$  activity peaks, the sedimentation rates resulting from the  $^{210}\text{Pb}$ -constant flux/constant sedimentation model,  $^{14}\text{C}$  ages and magnetic features. These data are from Bajard et al. (2016) for Lake La

Thuile, Wilhelm et al. (2012b) for Lake Blanc Belledonne, Wilhelm et al. (2012a) for Lake Allos and this study for Lakes Eychauda and Doménon. Age–depth models for cores were produced with the “clam” R-code package (Blaauw 2010). The interpolation between the dating points was made using a smooth spline function available in “clam”

(10–30%), as often observed in palaeolimnological studies of the last millennium because of the  $^{14}\text{C}$  age uncertainties.

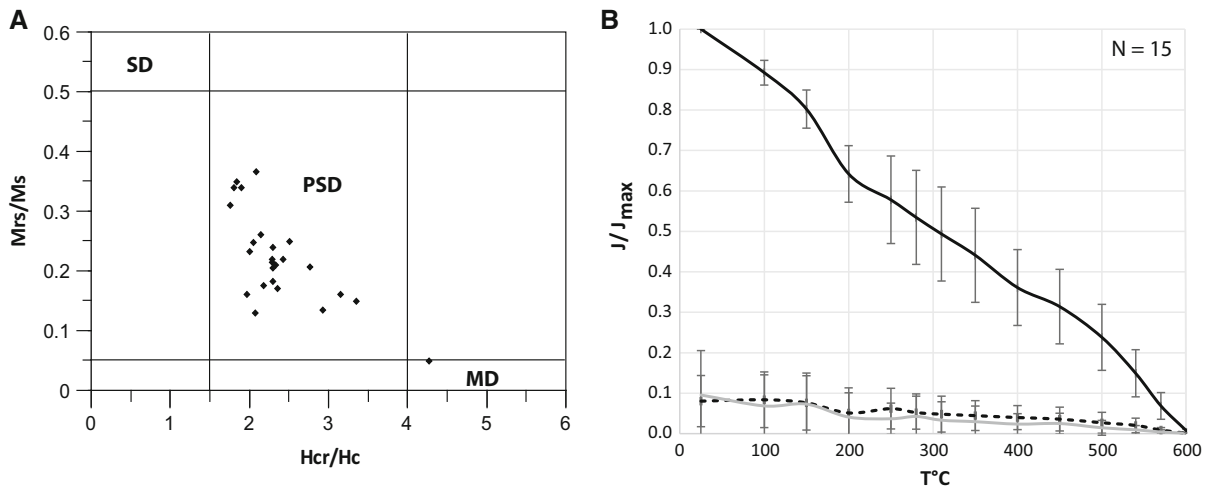
## Sediment magnetism and palaeomagnetism

### Magnetic mineralogy and magnetic properties

For the composite sequence of Lake La Thuile, 25 samples were analysed to retrieve the following hysteresis parameters: saturation magnetization, saturation remanence and coercivity. These parameters were determined from the paramagnetic-corrected hysteresis data. As illustrated in Fig. 5a, the saturation remanence divided by saturation magnetization and coercivity of remanence divided by coercivity are typical for pseudo-single domain magnetite according

to Dunlop (2002). The susceptibility versus temperature heating curves exhibit a clear peak in susceptibility between 420 and 560 °C indicating the neo-formation of magnetite, probably from the destruction of clay containing iron and/or sulphides. No change was observed around 300 °C. Therefore the occurrence of magnetic iron sulphides is not highlighted. The 3-axis Isothermal Remanent Magnetization (Fig. 5b) shows that the main carrier of the remanence is magnetically soft minerals with saturation mainly acquired between 0.1 and 0.3 T. The thermal demagnetization shows that all the Isothermal Remanent Magnetization (IRM) components are completely destroyed by heating at 600 °C, indicating that magnetite is the carrier of the remanence.

The magnetic parameters versus depth of the Lake La Thuile sedimentary sequence were studied

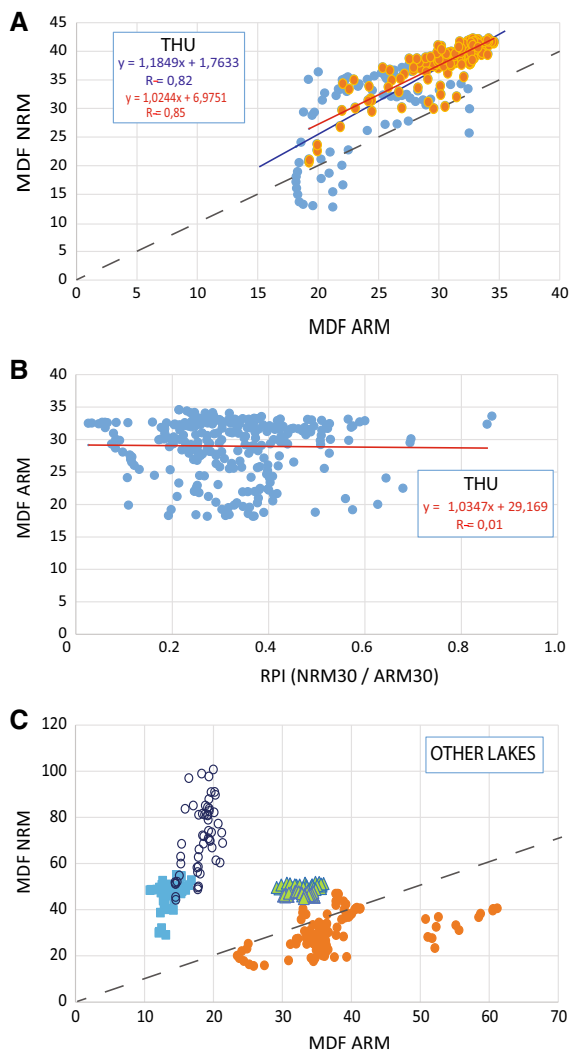


**Fig. 5** **a** Day diagram from hysteresis data for Lake La Thuile showing PSD grain size. *SD* single-domain grains, *PSD* pseudo-single domain grains, *MD* multi-domain grains. **b** Thermal demagnetization of 3-axis IRM (average of 15 samples). Grey,

black and dashed lines are Isothermal Remanent Magnetizations acquired in 1, 0.3 and 0.1 Tesla fields, respectively. See explanations in the text

(ESM2). The low field susceptibility, ARM40/ARM and ARM divided by the low field susceptibility show important changes at approximately 1.0 m depth. A negative correlation is observed between the low field susceptibility and the two other parameters, implying that they are mainly linked with grain-size changes. According to the age model (Fig. 4), the period of deposition of sediments located at 1 m deep corresponds to the Little Ice Age, therefore, these changes may be associated with an increase of terrigenous inputs as suggested by Bajard et al. (2016). In addition, the plot of the low field susceptibility versus Anhyretic Remanent Magnetization (ARM) indicates that the magnetic grain size ranges from 0.1 to 5  $\mu\text{m}$  (King et al. 1983). This observation indicates that the grain size is well constrained in the pseudo single domain range, suggesting no or few bacterial magnetite grains (Roberts et al. 2011). The S-ratio varies from 0.84 to 0.97 with an average of  $0.91 \pm 0.02$ . The median destructive field of the ARM varies from 18 to 35 mT with an average value of  $28.9 \pm 4.4$  mT, while the average value of the NRM's median destructive field is  $35.9 \pm 6.3$  mT. Comparison between the ARM and NRM median destructive fields (Fig. 6a) provides good correlation coefficients ( $r = 0.82$  and  $r = 0.85$  for the part below 1.0 m) and  $p$  values  $< 2.2e-16$ . The use of this comparison is allowed by the unidirectional behaviour of the characteristic remanent magnetization (ChRM)

illustrated in Fig. 3. The NRM is carried by the same particles as the ARM, given that the cross-correlation between the two parameters shows a slope close to 1. Moreover, the relative palaeointensity (NRM after 30 mT AF cleaning/ARM after 30 mT AF cleaning) which allows to be free from the effect of variations of low coercivity minerals concentration is independent of the grain size (Fig. 6b). Then, while relatively low values of the S-ratio demonstrate the occurrence of a high-coercivity phase, the other parameters suggest that pseudo-single domain magnetite grains of low-coercivity magnetization are the dominant carrier of remanence. Above a depth of 2.8 m, the ARM median destructive field and the ratio ARM/Saturated IRM are well correlated, suggesting that changes are linked to magnetic grain size variations. The decrease of the ARM/Saturated IRM ratio with the increase of low field susceptibility at approximately 0.5 m is interpreted as a grain size effect rather than a mineralogical change. Below 2.8 m, ARM/Saturated IRM ratios do not vary much, suggesting that the dominant magnetic mineral and grain size do not change significantly. The ARM/Saturated IRM would show higher values if the magnetic mineral were biogenic single domain magnetite in chains. The observed lower ratio might suggest that the magnetic mineral present in the sediment is not single domain magnetite. This observation confirms the results of hysteresis data highlighted in Fig. 5.



**Fig. 6** Illustration of the magnetic mineralogy behaviour in the sedimentary record. Cross-correlations between **a** the median destructive field (MDF) of natural remanent magnetization (NRM) and the median destructive field of anhysteretic remanent magnetization (ARM) for Lake La Thuile. **b** the relative palaeointensity (RPI) as NRM after 30 mT AF cleaning/ARM after 30 mT AF cleaning of Lake La Thuile. **c** The median destructive field of NRM and the median destructive field of ARM for the other lakes. Blue squares: Lake Blanc Belledonne, orange circles: Lake Allos, green triangles: Lake Doménon, open circles: Lake Eychauda. See explanations in the text

For the other lakes, the magnetic properties are summarized in Fig. 6c. For Lakes Doménon and Blanc Belledonne, the data are well grouped, indicating a stable magnetic mineralogy. For Lake Eychauda, the median destructive field of the Natural Remanent Magnetization (NRM) is very high (above 50 mT),

suggesting that the NRM is carried by a magnetic phase of high coercivity. Except for Lake Eychauda, the S-ratios indicate mainly magnetic phases of low coercivity. The low field susceptibility variations are usually associated with variations in the concentration of magnetic particles, clearly in relation with detrital inputs. For Eychauda, the low field susceptibility does not vary, suggesting quiet deposition or continuous detrital input. Variations of the low field susceptibility are negatively correlated to variations of the ratio ARM/low field susceptibility, demonstrating that magnetic properties are mainly linked to grain size and not to mineralogy. The other environmental magnetic parameters (i.e., ARM/Saturated IRM; ARM after 40 mT AF cleaning/ARM; median destructive field) are stable through each core, indicating that no major changes in the magnetic content occurred during sedimentation. The strongest variations in magnetic parameters are associated with grain-size changes and may reflect small amounts of haematite usually correlated to sedimentary detrital inputs. Thus, the main carrier of remanence in the sediments from the studied lakes seems to be pseudo-single domain magnetite.

The results of anisotropy of magnetic susceptibility measurements show well-preserved sedimentary features with the Kmin axis close to the vertical (i.e., parallel to the bedding pole). This observation suggests that no destruction of the sediment by coring, subsampling or bioturbation occurred.

#### *Characteristic remanent magnetization (ChRM) versus depth*

In most cases, the calculated ChRM is straight to the origin of the orthogonal diagram, highlighting only a single, highly stable magnetic component (Fig. 3). Occasionally, a viscous remanent magnetization overlaps the main component. This viscous remanent magnetization is removed by a weak alternating field (AF), usually 10 to 20 mT.

To visualize the results, a ChRM versus depth is built. For practical use, each ChRM versus depth sequence has been corrected to place the measured declination towards the northern direction. For Lake La Thuile, as the five sections are partly overlapping, a composite sequence is built as described above. Then, a second correction has been performed to superimpose the declinations measured in overlapping parts.

To avoid small gaps or artefacts, the declination and inclination data are smoothed with a moving average of 15 points width (i.e., 30 cm). For the other lakes, a sliding average over 10 cm depth is used to facilitate comparisons between them and with the reference curve.

In a few cases, breakdown of the sediment, either natural or related to the sampling mode (rotation, shearing, compaction or slugging in the corer or U-channel), have been evidenced by the scatter direction of the remanence. Measurement points with anomalous or inconsistent inclination have been deleted. The parts showing progressive shearing (i.e., trend in the variation of declination) through several decimetres have been either deleted or restored using a linear regression when possible. Such internal rotations have been interpreted by Ali et al. (1999) as due to spinning of the core barrel as it penetrates the sediment.

The inclination is often affected by various classic problems of compaction (Jackson et al. 1991, Colombat et al. 1993) and depositional mechanisms such as currents or floods (Garcés et al. 1996; Rösler and Appel 1998). The observed discrepancy may also be attributed to coring compaction (during penetration) or stretching (during extraction) in cohesive fine-grained sediments.

## Discussion

### Preservation and distortion of the palaeosecular variation (PSV) record

The question of the PSV record and therefore the mechanism of detrital remanent magnetization acquisition has been recently underlined by Valet et al. (2017). The NRM of sediments is the sum of the magnetization acquired by the orientation of detrital magnetic grains when falling in the water column and the biogeochemical magnetization produced by the alignment of magnetosome chains (fossils of magnetotactic bacteria) or other chemical processes in the few first centimetres of sediments, i.e., mainly in the oxic-anoxic transition zone (Tarduno et al. 1998; Faivre and Schüller 2008).

To test whether PSV records are reliable, several magnetic experiments can be performed, such as first-order reversal curve analyses (Egli et al. 2010).

However, most of the studied sediments have an important water content and a low magnetization, two points that do not allow the use of a vibrating sample magnetometer to construct first-order reversal curve diagrams. In the studied cores, the magnetic grains are pseudo single domain, suggesting a weak interaction between them. In addition, the magnetic mineralogy is dominated by fine-grained magnetite. Chemical changes (Rowan et al. 2009; Roberts 2015) or biomagnetization (Kim et al. 2005; Roberts et al. 2011) are not necessarily problems in our study if they occur soon after deposition as the magnetization is then blocked. Instantaneous deposits such as floods, mass wasting deposits or earthquake-induced deposits have thickness less than 2 cm, and even if the magnetization is affected, the measurement method smooths these possible artefacts induced by these layers.

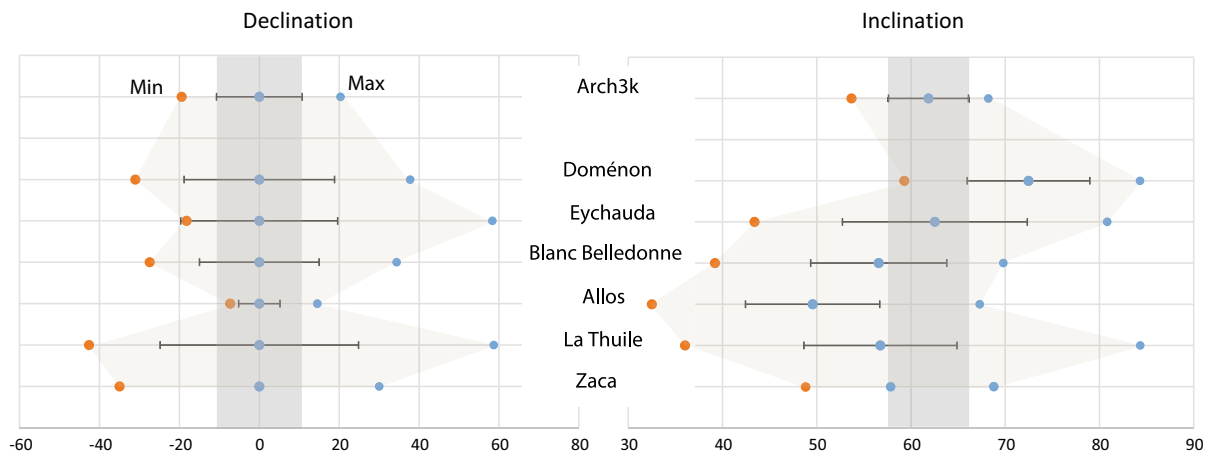
Beyond the magnetic mineralogy identification and sedimentological considerations, some of the poor results observed are related to possible deformation during coring or by bad recording processes.

Except for Lake Allos, the maximum angular variation (declination maximum – declination minimum) of the declination is significantly higher than that calculated for the Arch3k model (Table 2, Fig. 7). Similar discrepancies have also been observed in many palaeomagnetic records, for example, in Holocene western Canadian Arctic sediments with maximum angular variation of approximately 150° while

**Table 2** Standard deviation (STD) and maximal angular variation (MAV) of the declination and inclination of the Characteristic Remanent Magnetization for the different studied lakes compared to the Arch 3k model

	Declination		Inclination			Nb
	STD	MAV	Average	STD	MAV	
THU	24.8	101.3	56.8	8.1	48.3	218
ALO	5.2	21.8	49.6	7.1	34.8	64
BLB	15.0	61.8	56.6	7.2	30.6	49
EYC	19.6	76.5	62.5	9.8	37.4	46
DOS	18.8	68.8	72.5	6.5	25.0	33
ZACA	–	65.0	50.0	–	20	–
Arch 3k	10.7	39.7	61.9	4.2	14.5	160

The data from the lake Zaca (Lund and Platzman 2015) are indicated for comparison. The number (Nb) of values used for calculation is indicated



**Fig. 7** Minimum, mean and maximum angular deviation of declination and inclination for the studied lakes. Data from Zaca Lake (Lund and Platzman 2015) are centred around the theoretical inclination calculated according to the site latitude

the maximum angular variation of the reference curve is only approximately  $70^\circ$  (Barletta et al. 2010). The maximum angular variation of declination from Lake Zaca (California) is approximately  $65^\circ$  for the last 3 ky (Lund and Platzman 2015). A similar observation is possible in Huguangyan Lake (Yang et al. 2012), where the maximum angular variation of declination is twice that of the reference curve ( $60^\circ$  vs.  $30^\circ$ ). For Lakes Doménon, Blanc Belledonne and Eychauda, the maximum angular variations of declination are respectively  $68^\circ \pm 19^\circ$ ,  $62^\circ \pm 15^\circ$  and  $76^\circ \pm 20^\circ$ , less than twice those from Arch3k ( $40^\circ \pm 11^\circ$ ). For the Lake La Thuile composite section, the large variations ( $\sim 100^\circ$ ) can be partly explained by the bad quality of the overlaps between individual sections (Fig. 7).

According to the latitude of the sites and the expected range of secular variations for the last 2 ky, the inclination should vary from  $50^\circ$  to  $70^\circ$  (Fig. 7). Using the Arch 3k model as a reference, the inclination should vary from  $54^\circ$  to  $68^\circ$  with a mean and standard deviation of  $62^\circ \pm 4^\circ$ . The measured maximum angular variation of the inclination in the different lakes is approximately twice that from the Arch3k model. For Lakes La Thuile, Allos, Blanc Belledonne, Eychauda and Doménon, the mean and standard deviation of the measured inclinations are  $57^\circ \pm 8^\circ$ ,  $50^\circ \pm 7^\circ$ ,  $57^\circ \pm 7^\circ$ ,  $63^\circ \pm 10^\circ$  and  $72^\circ \pm 6^\circ$ , respectively (Table 2; Fig. 7). The measured values are thus low for Lake Allos and too high for Lake Doménon. For the other lakes, the measured values are in agreement with the expected values. The ChRM

inclination discrepancy can be due to sampling. However, it does not significantly affect the sedimentary structures, as evidenced by the well-preserved sedimentary anisotropy of magnetic susceptibility with the inclination of the minimum axis being close to the vertical.

The process of magnetization should also be questioned. The comparison between the median destructive fields of the Natural Remanent Magnetization (NRM) and the Anhyseretic Remanent Magnetization (ARM) (Fig. 6) allows testing for what types of magnetic grains carry the NRM. In the studied sediments, the NRM's median destructive field is higher than the one of the ARM, except for Lake Allos. For the other lakes, this observation can be explained by post-depositional processes or by a mixture with "haematite-like" grains. These grains are probably of detrital origin and come from the catchment. For Lakes Eychauda, Doménon and Blanc Belledonne, the catchment is dominated by crystalline rocks that with some alteration can produce haematite. For Lake Blanc Belledonne, the median destructive field of the NRM does not change, attesting that the magnetic grains carrying the NRM are very stable all along the core. For Lake La Thuile, most of the calculated median destructive fields of the NRM are well clustered at approximately 40 mT. The low values are mainly associated with high-susceptibility layers and coarse grains present in the upper part of the composite section. This feature may also explain the high maximum angular variation observed.

The use of paleo secular variation (PSV) as a complementary dating method is obviously dependent upon the nature of the sedimentary matrix and of the depositional processes. Undisturbed sediments are required, and such studies should thus avoid thick event layers such as mass slides, turbidites, flood deposits or bioturbated sediments. In the studied lakes, the sedimentary sequences are well laminated, suggesting that bioturbation is negligible. Additionally, for Lakes Eychauda, Blanc Belledonne and Allos, thin (< 1 cm) flood deposits do not seem to disturb the palaeomagnetic record as (1) measurements are realized each 2 cm and (2) each magnetic measurement point integrates approximately 4 cm width signal that smooths the effect of thin instantaneous deposits.

#### Comparison between cores and other lakes

For Lake Doménon, the general trends in declination and inclination variations with depth are similar between cores (DOS08-P2 and DOS08-P3, ESM3). In addition, the depth lags of the main declination and inclination changes are consistent with the stratigraphy. This observation leads us to be confident that the main features of directions versus depth are possible records of PSV. Similarly, for Lake Blanc Belledonne, the NRM of neighbouring cores BLB07P4, BLB0801 and BLB1204 exhibit similar trends of the inclination and declination curves, reinforcing the interpretation of a PSV record.

For cores ALO09P12 and EYC08P2, stable ChRM directions have been measured. Therefore, only one core was used. Then, to test validity of the PSV record, comparisons should be performed with neighbouring lakes (Fig. 8). In all the lakes, declination and inclination values are generally consistent between consecutive data measurements, revealing gradual directional changes. These changes appear to be very consistent from one lake to another (Fig. 8), suggesting a good record of PSV.

The Lake La Thuile composite sequence was compared to other PSV records in the western Alps (Fig. 9). Hogg (1978) produced PSV records covering the last 1500 years from Lakes Bourget and Annecy. For Lake Bourget and Lake Annecy, the stacked or smoothed curves available in the secular variation database from the IAGA Paleomagnetic Databases (<ftp://ftp.ngdc.noaa.gov/geomag/Paleomag/access>) were used. Good reproducibility is observed between the

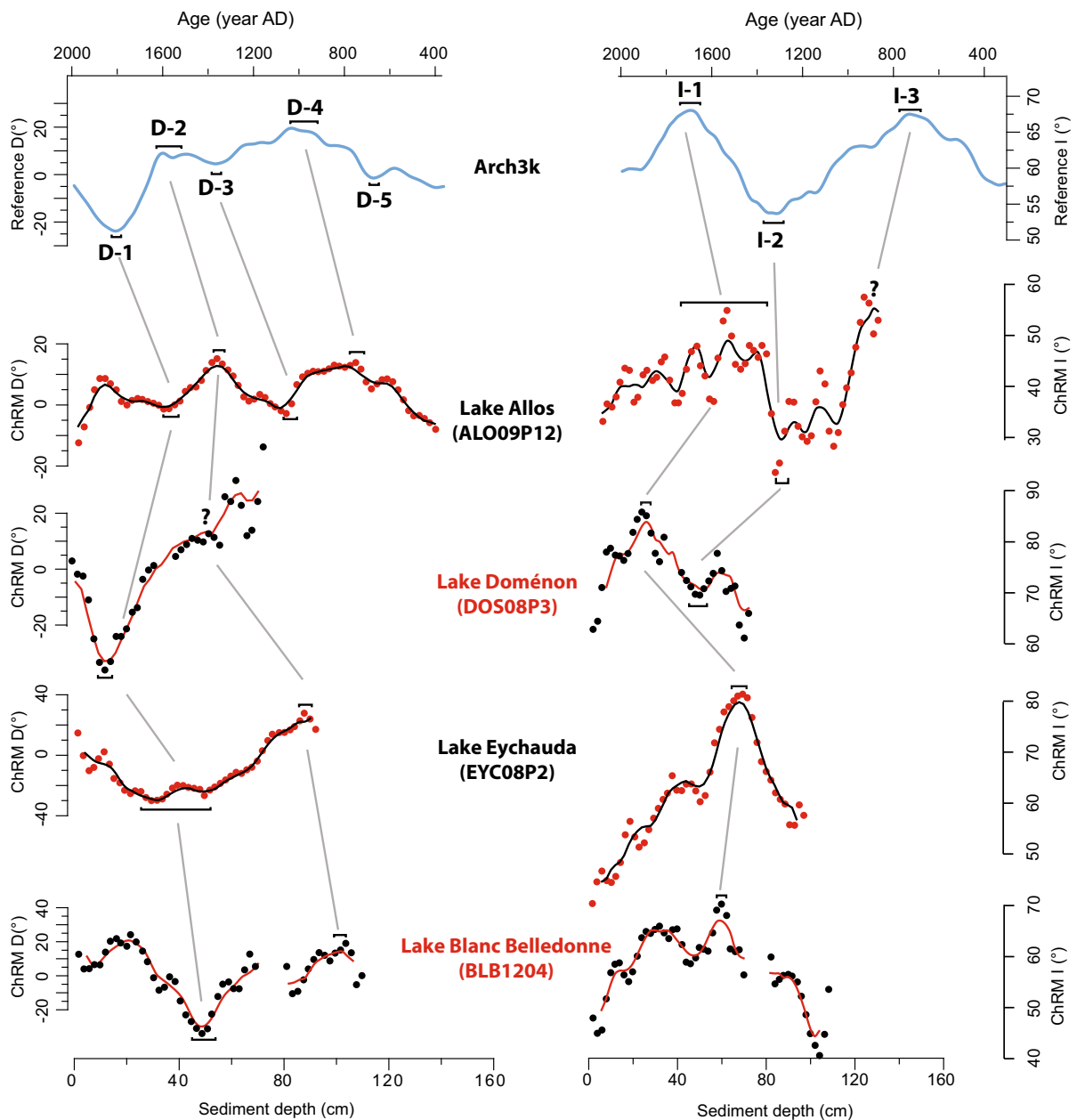
Lake La Thuile PSV and those from previously studied lakes. While maximal angular variations are high and magnetic mineralogy not perfectly stable all along the 5-m-thick section, this similarity suggests that La Thuile sediments may have recorded the PSV (Table 2).

For the different studied lakes, the changes in declination and inclination values are consistent (1) between neighbouring cores, (2) between different lakes and (3) with those from the reference curve (ARCH3k\_cst.1). All this evidence suggests a record of PSV to which is added some “noise”. Keeping in mind the possible problems, the declination/inclination versus depth curves can nevertheless be used to tentatively propose matching with the expected PSV reference curve to improve the age–depth relationships of the different lake records. For the best records (i.e., Lakes Blanc Belledonne and Allos), the correlation with the reference curve is efficient. For the other lakes, this correlation is more hazardous, and the comparison should consider other chronological constraints.

Relative palaeointensity is not used for correlation because the samples do not fulfil all the prerequisites that are needed for reliable intensity correlations, i.e., uniform grain sizes or mineralogy all along the section (Tauxe et al. 2010).

#### Age–depth model

An age–depth model cannot be produced by direct comparison between the Lake La Thuile paleo secular variations (PSV) and those of other lakes (Annecy, Bourget, Fig. 9) because of the poor quality of the chronology of the latter. These records are based on only a few  $^{14}\text{C}$  ages (Hogg 1978). Thus, a robust PSV reference curve is required to add magnetic chronological markers to the studied sediment records. The CALS10K reference curve is not used because it integrates many results from all around the world that are sometimes poorly constrained in time, such as those from Hogg (1978). As the ARCH3k\_cst.1 model uses only well-dated and selected archeomagnetic data mostly from Europe, it provides a more accurate high-resolution reference curve (Donadini et al. 2009 and Korte et al. 2009). A comparison of the Lake La Thuile PSV with the ARCH3k\_cst.1 model is illustrated Fig. 10. The declination profile shows, for instance, an ambiguous shape with several possibilities of



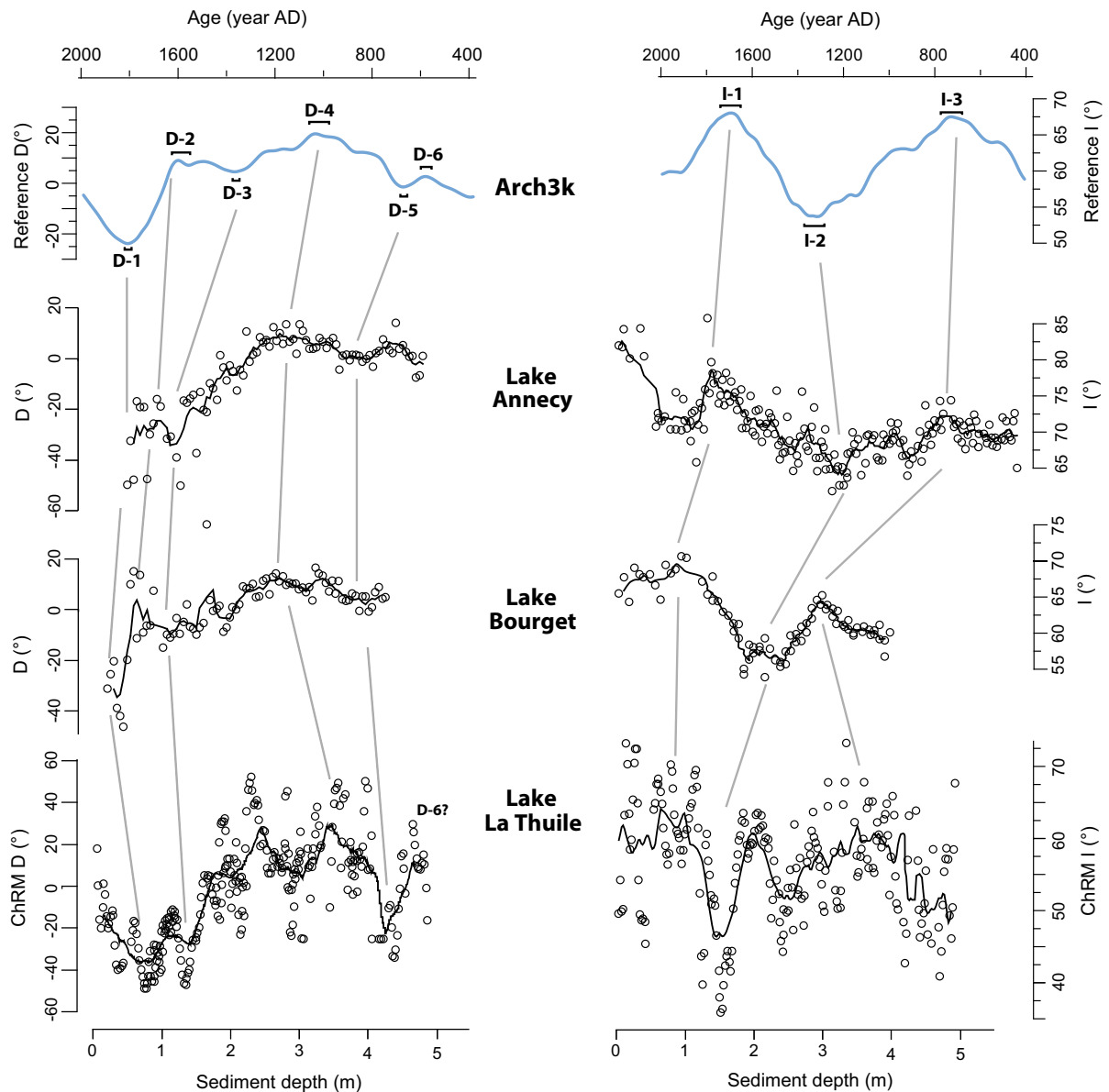
**Fig. 8** Correlative magnetic features (declination: D-1 to D-5 and inclination I-1 to I-3) between the expected geomagnetic model ARCH3k\_cst.1 (Korte et al. 2009) and the ChRM profiles

of cores EYC08P1, BLB0704, ALO09P12, BLB1204 and DOS08P3. Note that the hiatus in the BLB1204 curves is due to the presence of a coarse sand layer

correlation to the reference curve. The inclination profile and the  $^{14}\text{C}$  probability distribution help to select the most likely correlation. The third inclination (I-3) feature is not clearly observable, as the inclination profile does not show any obvious change below 2.5 m. Hence, only two inclination features (I-1 and I2) can be well defined and used for chronological

purposes. The ARCH3k\_cst.1 model reference curve shows well-defined peaks of declination (D-1 to D-5) and inclination (I1 to I3) that are usually easy to correlate with the PSV records from different lake (Figs. 7, 8).

For each studied lake, the comparison between Characteristic Remanent Magnetization (ChRM)



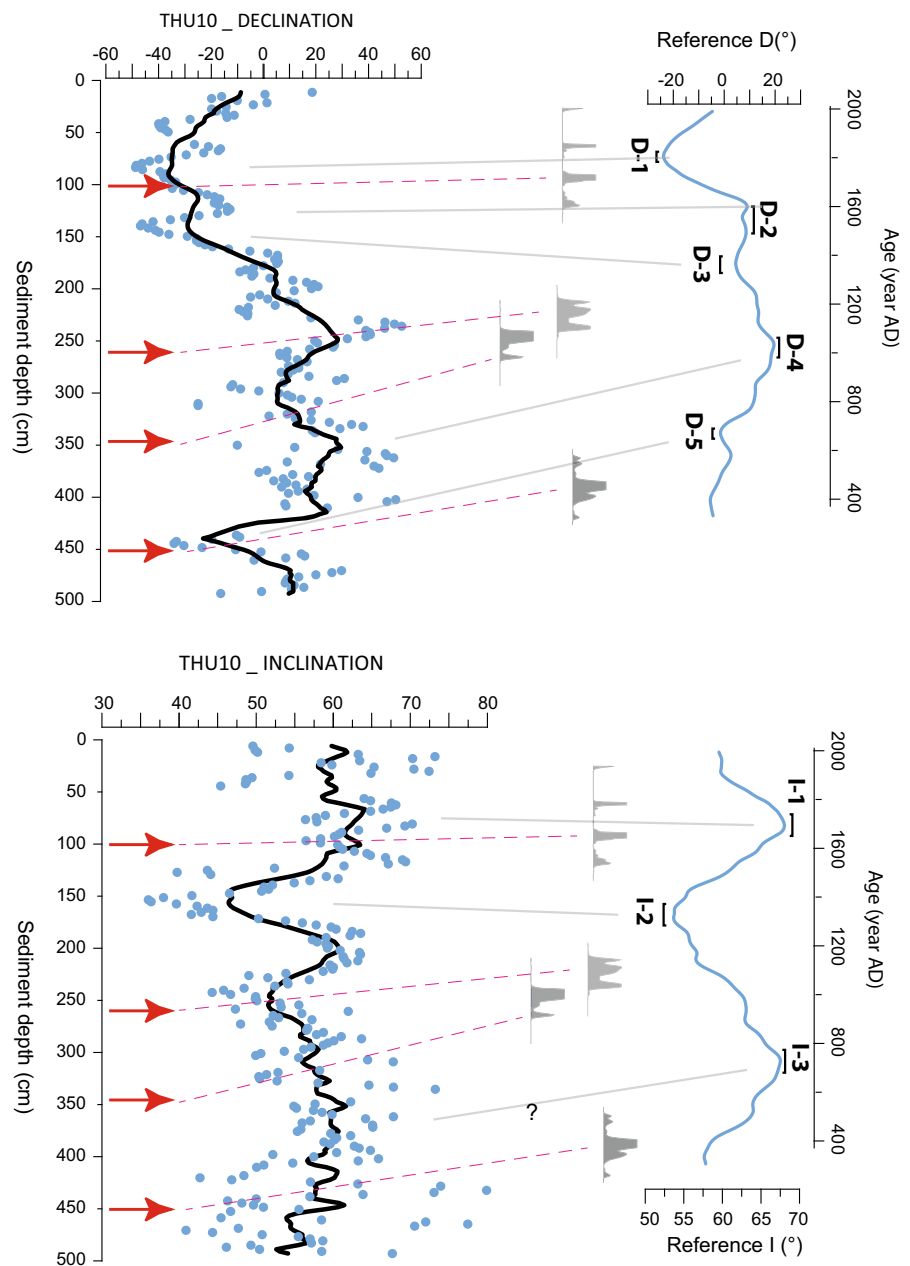
**Fig. 9** Correlation between Lake La Thuile, Lake Annecy and Lake Le Bourget (Hogg 1978) with the ARCH3k\_cst.1 model for the Characteristic Remanent Magnetization (ChRM)

versus sediment depth and the reference curve (ARCH3k\_cst.1) versus ages provides chronological markers. For the studied lakes, these correlations are usually simple (Figs. 8, 9, 10). In few cases, several correlations are theoretically possible. For example, the inclination curve of BLB1204 shows two peaks. Only the deepest (and most pronounced) is in agreement with the declination curve.

declination and inclination. The ages of D-1 to D-5 and I-1 to I-3 with uncertainty are determined according to ARCH3k\_cst.1 curves

These magnetic chronological markers are compared to other chronological data ( $^{14}\text{C}$  ages, short-lived radionuclide data and historical events) to improve the original age–depth relationships. According to the ARCH3k\_cst.1 model, the errors in age are estimated as correspondent at a variation of  $0.5^\circ$  around the D/I – x. The error bars in depth are

**Fig. 10** Comparison between the Characteristic Remanent Magnetization (ChRM) declination and inclination versus depth from core THU-10 from Lake La Thuile and the ARCH3k\_cst.1 model. Arrows indicate the depth of  $^{14}\text{C}$  samples, and their calibrated ages are next to the magnetic reference curve (in years AD)



determined visually according to the shape of the ChRM versus depth curves.

The proposed correlations ( $D/I - x$  vs. age), including error estimations, are consistent with other available chronological data (Fig. 4). That is, they provide a strong argument in favour of a consistent record of PSV. Moreover, the addition of these PSV chronological markers contributes to reduce mean uncertainty from 67 to 49 years, 130 to 91, 101 to 52

and 139 to 86 years for Lakes Blanc Belledonne, Doménon, Allos and La Thuile, respectively. Using the markers produces more than 27% uncertainty reduction, which allow more precise discussion of the timing of palaeoclimatic or palaeoenvironmental variations. For Lake Eychauda, the use of PSV allows establishment of an age–depth model with a regular shape, while no  $^{14}\text{C}$  macroremains were found initially, limiting the use of this sediment sequence.

## Conclusions

From a study of the sediment magnetic mineralogy and remanence of five alpine lakes in the French Alps (La Thuile, Allos, Eychauda, Blanc Belledonne and Doménon), we draw the following conclusions:

- The natural remanent magnetization is mainly carried by low-coercivity pseudo-single domain magnetite.
- The characteristic remanent magnetization direction is consistent at the site level between neighbouring lakes and with the reference curve, suggesting that secular variations of the geomagnetic field are approximately recorded. The maximum angular variations of the declination and inclination are used to highlight the discrepancies with the paleo secular variation model used.
- The acquisition of the remanent magnetization seems to be effective in the first few centimetres of sediment.
- The agreement of the records of palaeosecular variations with available independent chronological data for the different studied lakes supports the interpretation that sediments have recorded the palaeosecular variations more or less accurately depending on the studied lake.

Following these findings, marked changes in the palaeosecular variations have been tested as chronological markers in the age–depth modelling. Using these markers results in a reduction of dating uncertainties up to 27%. This result strongly supports that paleo secular variation may help to improve lake sediment chronologies. However, checking the reliability of the palaeomagnetic signal is a mandatory prerequisite, and this check should also be coupled with a comprehensive understanding of the sedimentary and magnetic processes through a multiproxy (sedimentology, geochemistry, dating) analysis.

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