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Detrital events and hydroclimate variability in the Romanian Carpathians during the mid-to-late Holocene

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ABSTRACT

The Romanian Carpathians are located at the confluence of three major atmospheric pressure fields: the North Atlantic, the Mediterranean and the Siberian. Despite its importance for understanding past human impact and climate change, high-resolution palaeoenvironmental reconstructions of Holocene hydroclimate variability, and in particular records of extreme precipitation events in the area, are rare. Here we present a 7500-year-long high-resolution record of past climatic change and human impact recorded in a peatbog from the Southern Carpathians, integrating palynological, geochemical and sedimentological proxies. Natural climate fluctuations appear to be dominant until 4500 years before present (yr BP), followed by increasing importance of human impact. Sedimentological and geochemical analyses document regular minerogenic deposition within the bog, linked to periods of high precipitation. Such minerogenic depositional events began 4000 yr BP, with increased depositional rates during the Medieval Warm Period (MWP), the Little Ice Age (LIA) and during periods of societal upheaval (e.g. the Roman conquest of Dacia). The timing of minerogenic events appears to indicate a teleconnection between major shifts in North Atlantic Oscillation (NAO) and hydroclimate variability in southeastern Europe, with increased minerogenic deposition correlating to low NAO index values. By linking the minerogenic deposition to precipitation variability, we state that this link persists throughout the mid-to-late Holocene.

1. Introduction

The Carpathian Mountains and bordering lowlands are one of the most rapidly reacting regions of Europe to current climatic change, with droughts, and periods of short, intense precipitation becoming more common (IPCC, 2014; Micu et al., 2015). The wider region (the Carpathian-Balkan) is located at the confluence of major atmospheric circulation patterns, with the North Atlantic system towards the west, the Mediterranean to the southwest, and the Siberian High to the east (Obreht et al., 2016 and references therein; Panagiotopoulos et al., 2005). As a result, the region should be very sensitive in recording past climate variability resulting from periodic shifts in the dominant circulation pattern. The North Atlantic Oscillation (NAO), in particular, has a major control on winter precipitation (Bojariu and Giorgi, 2005; Stefan et al., 2004; Tomozeiu et al., 2005, see Fig. 1C) and winter temperature (Bojariu and Giorgi, 2005) changes in the region.

The Carpathian-Balkan region is one of the longest-inhabited regions in Europe, with Neolithic cultures having interacted with the environment as far back as 9000 years before present (yr BP) (Bailey, 2000). An increasing number of studies have demonstrated the importance of the long-term impact of humans in the Carpathians, particularly via deforestation and high Alpine pasturing (Carozza et al., 2012; Feurdean et al., 2009; Feurdean and Astalos, 2005; Schumacher et al., 2016), activities which may have a significant impact on an area's erosional regime and sediment budget (e.g. Arnaud et al., 2012). Indeed, the Balkan Peninsula was the earliest region in Europe to domesticate animals, roughly 9000 yr BP (Larson et al., 2007), and hosted the spread of agriculture from the southeast from 7000 yr BP (Price, 2000; van Andel and Runnels, 1995). Additionally, the earliest known examples of extractive metallurgy (around 7000 yr BP) may be found throughout the region (Radivojević et al., 2010 and references therein); evidence for a long history of significant human impact.

Despite significant improvements in the last decades, high-

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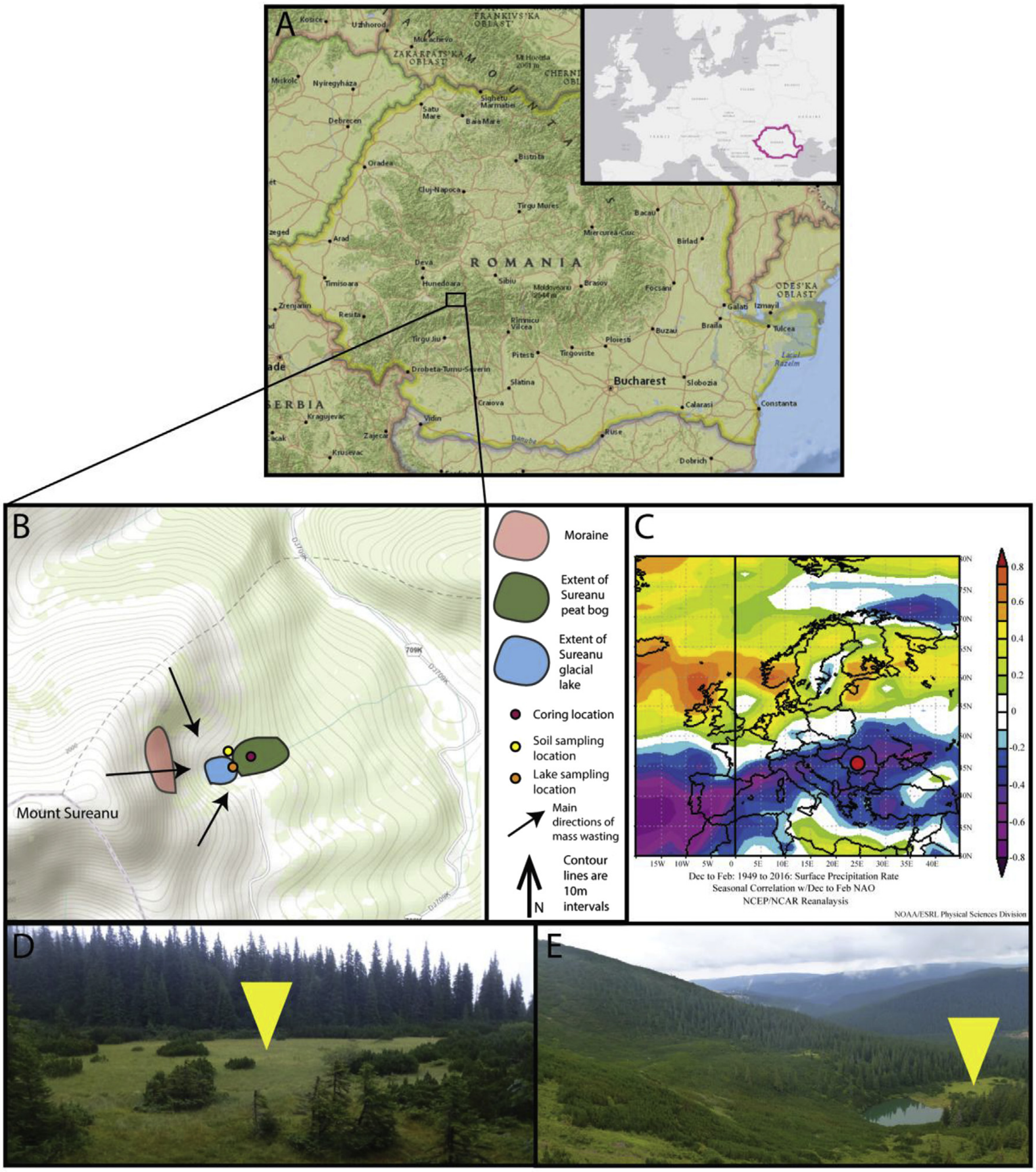

Fig. 1. Location of Sureanu bog (A). B: Topographical map indicating location of bog, lake (Iezerul Sureanu), moraine, all sampling sites and flow directions of mass wasting. C: Correlation of high North Atlantic Oscillation (NAO) index to winter precipitation, indicating reduced precipitation at times of intense NAO in Eastern Europe. Location in the Southern Carpathians denoted by red circle. D: Closer view of bog, indicating location of coring site (yellow arrow). E: View looking down onto site, with coring site indicated by yellow arrow. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

resolution and especially multi-proxy palaeoclimate records from the Carpathian region in Romania are still rare (Buczkó et al., 2013; Magyari et al., 2009, 2012, 2014; Haliuc et al., 2017). Individual proxies have been used to produce a number of long-term records, especially pollen (Feurdean et al., 2008a,b; Schumacher et al., 2016; Tanţău et al., 2011 and references therein), speleothems (Onac et al., 2002; Constantin et al., 2007; Drăguşin et al., 2014) and other palaeoecological and geochemical proxies (Brückner et al., 2010; Magyari et al., 2013; Schnitchen et al., 2006; Tóth et al., 2015). Most studies display strong inter-site variability, even when in close proximity to one another (e.g. Feurdean et al., 2008a,b), an indication of the complexity of the regional climate, one which has been defined primarily by natural controls (e.g. Tóth et al., 2015). In addition, the environment has been heavily influenced by major anthropogenic disturbances (Schumacher et al., 2016). Furthermore, a tree ring reconstruction of summer temperatures over the past 1000 years in the Eastern Carpathians (Popa and Kern, 2009) shows an interesting lack of correlation to similar records from central Europe (e.g. Büntgen et al., 2011a,b), particularly during periods of rapid climate change (Medieval Warm Period (MWP), Little Ice Age (LIA)). This is indicative of strong regional forcing of climate in the Romanian Carpathians in particular and south-eastern Europe in general (Roberts et al., 2012). This is further evidenced by pollen-based reconstructions across the continent, which indicate a disconnection between Holocene temperature and precipitation changes in south-eastern Europe, and central and western Europe (Davis et al., 2003; Magny et al., 2013; Mauri et al., 2015). The location of the region, at the confluence of three major atmospheric pressure fields, may play a major role in this apparent discrepancy. Studies exploring this teleconnection between changes in the atmospheric system and the impact on the environment are rare in this area (Haliuc et al., 2017), as most studies have been focussed on the last 100 years (e.g. Bojariu and Giorgi, 2005 for a review). These studies indicate the correlation of a low NAO index with high precipitation in the region for the period of available meteorological data (Stefan et al., 2004; Tomozeiu et al., 2005; see Fig. 1C), but it is unclear if this connection has persisted over a longer timescale at a regional scale.

To understand the link between changing atmospheric patterns and precipitation, records of sedimentation related to flooding events, common in central Europe, may be used (Czymzik et al., 2013; Magny et al., 2013; Swierczynski et al., 2013; Wirth et al., 2013). Using these long term high-resolution records in central Europe, a link between NAO variability and periods of flooding has been inferred (Wirth et al., 2013).

Determining the interactions between varying controls on the climate system is difficult when utilising single-proxy studies, particularly when attempting to put a region's history in the context of a changing climate and human occupation, and so more multi-proxy studies are needed (Veres and Mîndrescu, 2013). Here we present the first record of apparent flooding events from Southeastern Europe, in the Southern Romanian Carpathians throughout the mid-to-late Holocene. Alongside proxies for organic matter and minerogenic contents, we investigate the viability of a novel geochemical approach (namely the Rb/Sr ratio) as a proxy for minerogenic deposition in the bog, previously only utilised in loess and lake sediments (Jin et al., 2006; Vasskog et al., 2011).

2. Regional and local setting

Sureanu peat bog (45°34′51″N, 23°30′28″E), is a small bog located adjacent to a tarn (Iezerul Sureanu) in the Sureanu Mountains, Southern Carpathians (Romania) at an elevation of 1840 m above sea level (a.s.l) (Fig. 1). Iezerul Sureanu is roughly 100 m long, and 90 m wide, with a maximum depth of 7.5 m. It is frozen for around 6 months a year, and is separated from the bog basin by a morphological rise, likely a small moraine (See Fig. 1 and SI 1). Further upslope is another palaeomoraine, with exposed rock and possible avalanche channels. Other mass-wasting related features may be masked by the dense vegetation (See Fig. 1D). The bog itself is roughly 200 m long and 100 m wide, and is surrounded on three sides by the steep slopes of the Sureanu palaeoglacier cirque, with a slope gradient in excess of 1 in 2 (see Fig. 1). The bog is slightly raised, with occasional streams at its extremities that periodically drain the Iezerul Sureanu Lake during high water stands. The bog vegetation is dominated by *Sphagnum*, with patches of *Lycopodium*, and various species of Poaceae and Cyperaceae. The peatbog is still uncovered, with peripheral forests consisting primarily of *Picea abies* (spruce) and *Alnus glutinosa* (alder). At lower altitudes, *Fagus sylvatica* (beech) occurs, a vegetation assemblage typical of the *Picea* belt of the Carpathian Mountains. The location of the bog, at the uppermost extent of the spruce forest in this area (circa 1800m, as defined by Cristea, 1993), and just below the transition into the subalpine belt (dominated by *Pinus mugo* and *Juniperus*) means the bog should be well placed to capture shifts in vegetation. Due to its location, the bog is likely to preserve a record of periods of high precipitation through the associated land erosion and minerogenic deposition. This is because it sits at the base of steep slopes, and is hydrologically coupled to the neighbouring Iezerul Sureanu Lake, and so should receive input of sediment from mass wasting of the slopes, or flooding of the lake, although parts of the runoff will be deposited in the lake (when it is not frozen).

The climate of the area is considered as temperate continental (Fărcaş and Sorocovschi, 1992) with average winter temperatures ranging from −2 °C below and −7 °C above 1900m a.s.l., and corresponding average summer temperatures of 19 °C and 8 °C respectively at the same elevations.

Due to the interplay of Mediterranean and Atlantic air masses, temperature inversions are common, with resultant fluctuations especially prevalent in the winter and spring (Trufas, 1986). Rainfall amounts are between 900 and 1800 mm per year, with extensive snow cover common throughout the winter (around 100 days a year at low altitudes and over 200 days above 2000 m a.s.l.). In common to other regions in the Southern Carpathians, the area receives precipitation mainly of Atlantic origin, but with periodic incursions of south-easterly air masses from the Mediterranean Sea (Fărcaş and Sorocovschi, 1992).

The basement geology is dominated by Late Proterozoic – Early Paleozoic gneisses of the Getic-Supragetic nappe (Iancu et al., 2005). Recent human impact can be seen in the form of ski slopes constructed on the far side of Sureanu peak (See Fig. 1B) and recent deforestation to allow for building of hotels in the area, as well as long-term (centuries if not millennia old) high-altitude pasturing and hay harvesting on the high plateau of Sureanu Mountains.

3. Materials and methods

3.1. Coring

A 603 cm long core was taken in October 2014 using a Russian peat corer (diameter 5 cm) in the central part of the bog, in 100 cm long sections. Two parallel and overlapping cores were taken for every depth to ensure the entire peat record was recovered. Additional samples were taken from local rocks, lake sediment and soil, to allow for characterisation of debris found within the bog record. One sediment sample consisting of fine gravel and sand was taken from the lake-shore, and a soil sample from the surrounding vegetated slopes between lake and bog. Both were sampled by hand trowel, down to a depth of 5 cm. The cores were wrapped in cling film before transportation to Northumbria University, where they were kept at 3 °C prior to analysis. The core was documented and described prior to being photographed in the lab. It was then cut into 1 cm slices prior to selection of samples for future analysis.

3.2. Loss-on-Ignition

Loss-on-ignition (LOI) was performed on 1 g of wet sediment, with samples taken every centimetre, dried overnight (at 105 °C), prior to initial ignition at 550 °C for four hours on samples from every centimetre. Weight loss after combustion at 550 °C can be used to calculate combusted organic material (Heiri et al., 2001), prior to combustion at 950 °C for a further two hours to gain total carbon content through the removal of carbonates. What remains after such combustion is the minerogenic matter (MM), with what has been combusted the organic matter (OM) fraction.

3.3. Geochemistry

For trace element analysis, 0.2 g of dried, homogenized sediment was taken for each analysed sample. Each sample underwent a mixed acid digestion, using HNO_3 (9 ml), HCl (3 ml) and HF (0.5 ml) (method adapted from Krachler et al., 2002) and a MARS microwave accelerated reaction system, with a 40-min pressurised heating phase followed by 20 min of cooling. This produced clear digests with minimal residue when diluted to 50 ml with mili-Q deionized water. Aliquots of the sample (9 ml) were taken, alongside 1 ml of 100 ppm Spex CertiPrep Yttrium internal standard, before analysis via a Perkin Elmer Optima 8000 ICP-OES at Northumbria University. For the analysis of Rb, Sc and Sr, wavelengths of 780.023, 407.771 and 361.383 were used, respectively.

A calibration standard (Aristar ICP-MS Calibration Standard 2, at 0.01, 0.1, 1, 2, 5, 10 and 20 mg/L) was run prior to each sample run to produce a calibration curve. Alongside the samples, acid blanks and two standards were run, Montana 2711 soil and IAEA-SL-1 Lake Sediment which indicate that most values fall within 10% of expected (see Table 1, and SI 2), with only Rb showing slightly higher recovery. Blanks were shown to be negligible, indicating no outside influence on the method.

3.4. Palynological analysis

Samples of 1 cm^3 sediment were taken throughout the core at roughly 10 cm intervals, with a total of 65 samples counted. These were prepared following standard palynological methods with acetolysis and hydrofluoric acid digestion (Faegri and Iversen, 1989). *Lycopodium* marker spores were added, to allow for concentrations to be calculated (Stockmarr, 1971). At least 250 grains (excluding those not included in the pollen sum; fungi, aquatics and unidentified grains) were counted in all samples, and pollen percentages were produced from the number of counts respective to the total pollen sum, not including aquatics, fungi, and unknown grains (of which there were less than 3 per sample). When presented, pollen percentages for aquatics were calculated from a separate pollen sum including aquatic taxa. Pollen and spores have been identified using literature (Beug, 2004; Demske et al., 2013) and the pollen reference collection held at Northumbria University. Graphs were drawn using the software Tilia (Grimm, 1990). Microscopic charcoal content was calculated using the point count method as outlined by Clark (1982). Local pollen assemblage zones (LPAZ) were delimited by stratigraphically constrained cluster analysis in CONISS (Grimm, 1987).

3.5. Chronology

The age model for the core is based on 19 14C dates. With one exception (sample DeA-5795 consisting of wood), all bulk sediment samples submitted to dating consisted mainly of moss fragments (Table 2). Radiocarbon dating was performed via accelerator mass spectrometry (AMS) at the ^{14}C CHRONO centre at Queen's University Belfast, and at HEKAL AMS Laboratory, MTA ATOMKI Institute for Nuclear Research of the Hungarian Academy of Sciences in Debrecen (Molnár et al., 2013). The ^{14}C ages were converted into calendar years using the IntCal13 calibration curve (Reimer et al., 2013) and an age-depth model was generated using Bacon (Blaauw and Christen, 2011, Fig. 2). Sample DeA-5795 represents an outlier and therefore was excluded from our age modelling. This is a wooden sample, which may be much older than surrounding sediment and was likely transported onto the bog from the surrounding slopes (Oswald et al., 2005; Schiffer, 1986).

3.6. Grain size

For granulometric analyses, approximately 1 g of sample was taken, prior to removal of the organic fraction through the addition of 15 ml H_2O_2. Samples were allowed to settle for 2 h before heating on a hotplate at 200 °C to dry down the remaining non-organic fraction. These dry samples were then resuspended in 40% Calgon using a sonicator before analysis via a Malvern Mastersizer Particle Size Analyser. Results were analysed and presented using Mastersizer software prior to interpretation. Such analysis cannot indicate the presence of grains >2 mm, and so the very largest grains are not indicated.

4. Results

4.1. Lithology, sedimentology and age model

The lower part (below 300 cm) of the record consists of gyttja and fen peat, clearly documenting the gradual transformation of this basin into a raised bog throughout the mid-to-late Holocene (See Fig. 3). For the upper 4 m, the record consists of *Sphagnum*-dominated peat, with a number of minerogenic layers, some of which are apparent upon visual inspection.

The age model (Fig. 2) indicates the time frame covered by this study is from roughly 7500 yr BP to the present day, with the uppermost peat (growing moss) dating from 2014. This means the time span between samples is roughly 10yrs for the LOI and grain size, 100yrs for the pollen and between 30 and 100yrs for the geochemistry. All ages quoted throughout the text are in calendar years BP (yr BP).

For the first 2000 years, the Sureanu record is characterised by high minerogenic matter (MM) values (roughly 70%), indicative of a coarse gyttja-like, lacustrine sediment with occasional roots and fine gravel (Fig. 3). After 5000 yr BP there is a shift towards a coarse, detritus-rich fen peat, with MM values stable around 40–50%, but with small high-organic excursions becoming more common throughout the period, pointing to the gradual establishment of a raised bog. There is a clear transition into much more detritus-free peat at around 3300 yr BP, which follows on from a shift in the MM record to values of around 90% at 3500 yr BP. There are occasional sharp rises in the MM values however, but it is not until 2140 yr BP these become more frequent. In this section, the record consists of fine *Sphagnum* peat, with clear occasional root and sand/small pebbles incursions. This period is characterised by the first appearance of regular, sharp (in terms of boundaries) and rapid (in

Table 1
Certified Reference Material (CRM) recoveries for elements analysed via ICP-OES. Values of expected and observed concentration for the selected elements are presented, along with the average recovery for each element for the two CRMs. For both CRMs, 5 replicates were run throughout the analyses.

Element	Expected (ppm)	Observed Average (ppm)	Average Recovery (%)	Standard Deviation
Montana Soil 2711				
Rb	110	127.5	115.9	17.73
Sr	245.3	221.4	90.2	14.79
Sc	9	8.9	99	1.38
IAEA Lake Sediment				
Rb	113	124.6	110.2	9.53
Sr	80	95.5	119.3	4.26
Sc	17.3	17.5	101.1	1.04

Table 2
Radiocarbon dates used to build age model for Sureanu record. Sample DeA-5795, dated on wood, is likely an age outlier and was excluded from age model calculations.

Lab No.	Depth	^{14}C age (±1σ)	Minimum Calibrated Age (yr BP)	Maximum Calibrated Age (yr BP)	Mean Calibrated Age (yr BP)	Dated material	Observation
DeA-7256	34	−16 ± 22				bulk peat	
DeA-7257	57	236 ± 23	277	310	287	bulk peat	
DeA-7258	72	368 ± 22	319	500	441	bulk peat	
DeA-7259	104	805 ± 22	680	770	713	bulk peat	
DeA-7260	129	728 ± 21	660	690	675	bulk peat	
DeA-7261	163	1164 ± 25	989	1177	1090	bulk peat	
DeA-5795	172.5	3176 ± 26	3360	3450	3401	wood	outlier
DeA-7262	200	1417 ± 26	1290	1356	1319	bulk peat	
DeA-5796	209	1560 ± 27	1392	1528	1467	bulk peat	
DeA-7263	228	1809 ± 22	1634	1820	1750	bulk peat	
UBA-31373	252	2228 ± 44	2148	2339	2244	bulk peat	
UBA-31374	280	2202 ± 44	2119	2333	2226	bulk peat	
DeA-7264	314	2595 ± 24	2720	2759	2744	bulk peat	
UBA-31375	344	3023 ± 30	3141	3275	3207	bulk peat	
UBA-31376	378	3317 ± 50	3447	3645	3551	bulk peat	
DeA-7265	404	3638 ± 29	3868	4080	3949	bulk peat	
UBA-31377	454	4181 ± 32	4614	4706	4660	bulk peat	
DeA-5797	516	5301 ± 36	5950	6189	6083	bulk peat	
UBA-31378	603	6777 ± 54	7564	7702	7633	bulk peat	

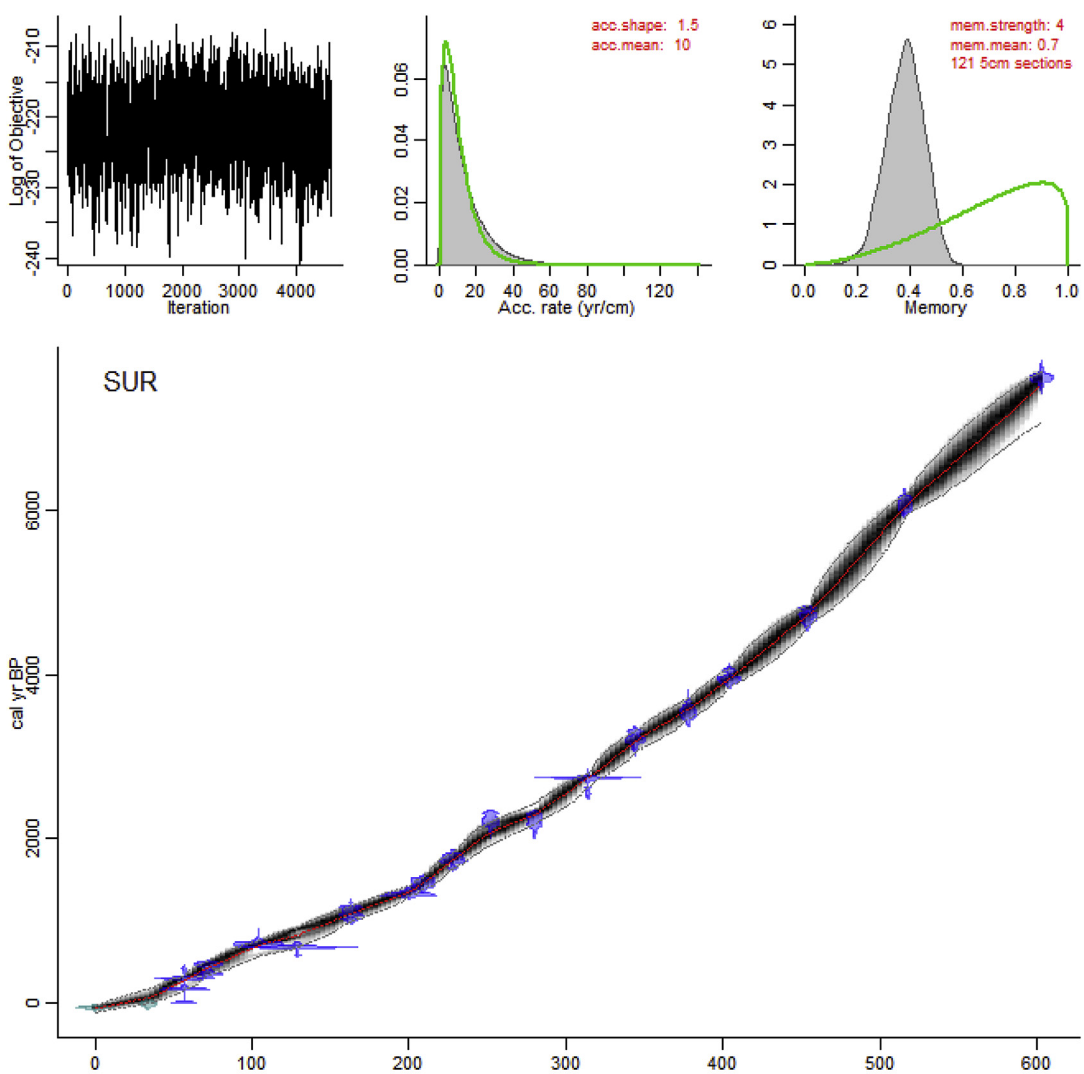

Fig. 2. Age model for Sureanu record based on 18 ^{14}C dates, as calculated using Bacon (Blaauw and Christen, 2011). Upper left graph indicates Markov Chain Monte Carlo iterations. Also on the upper panel are prior (green line) and posterior (grey histogram) distributions for the accumulation rate (middle) and memory (right). For the lower panel, calibrated radiocarbon ages are in blue. The age-depth model is outlined in grey, with darker grey indicating more likely calendar ages. Grey stippled lines show 95% confidence intervals, and the red curve indicates the single 'best' model used in this work. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

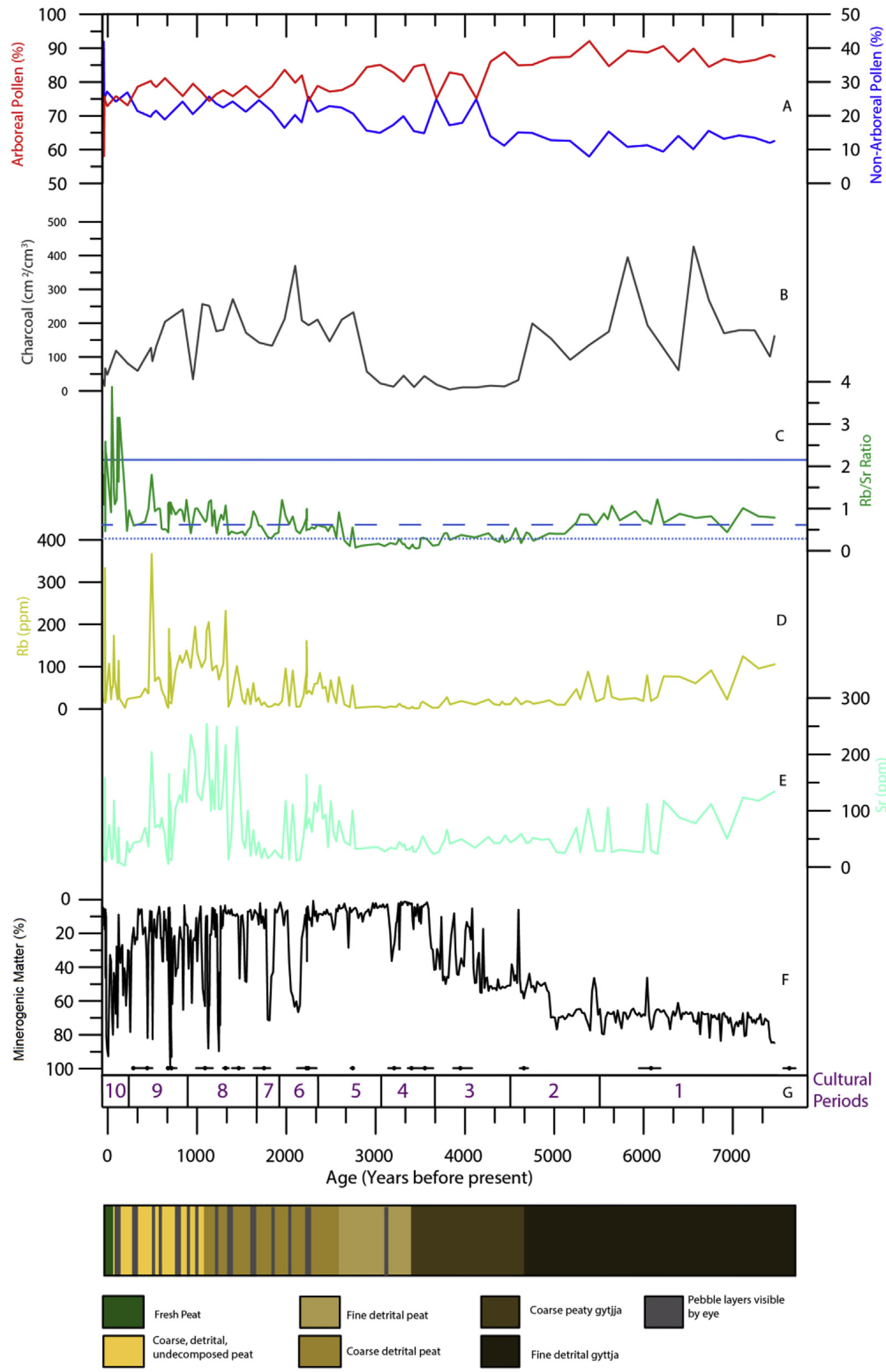

Fig. 3. Downcore variations in multiple proxies from Sureanu. Arboreal and non-arboreal pollen percentages (A) are presented alongside microcharcoal counts (B), the Rb/Sr ratio (C), lithogenic (Rb, Sr) element concentrations (D and E). The blue lines in panel C denote the ratio from local soil (solid), lake sediment (dashed) and local rock (dotted). Panel F shows the minerogenic matter as determined via loss-on-ignition. Cultural periods referenced in the text are indicated in panel G: 1: Neolithic, 2: Early Bronze Age, 3: Middle Bronze Age, 4: Late Bronze Age, 5: Iron Age, 6: Dacian State, 7: Roman Dacia, 8: Middle Ages, 9: Medieval Period, 10: Industrial to modern. Calibrated radiocarbon dates and uncertainties are indicated at the base of the graph. In addition, a simple lithological diagram is presented. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

terms of deposition time) minerogenic depositional events within the peat. The first of these is seen in the MM record between 2140 and 2030 yr BP, with an excursion to MM values as high as 70%, before a short period of normal bog growth and subsequent minerogenic overprinting again around 1870 yr BP. This large event is followed by the final period of extended uninterrupted peat deposition, with MM values of 10% sustained until 1260 yr BP (Fig. 3F).

After 1260 yr BP there are 13 major high minerogenic content depositional events (see Fig. 3). These events are variable in their duration and their presumed effect on the bog growth. Most cover a fairly short period of time (roughly 5–10 years or even less), but show MM values as high as 90%. Some indicate MM of around 60% and last a little longer. A period of constant high MM is observed between 1100 and 850 yr BP, in which the values rarely fall below 20%. The sediment remains a detrital peat (still composed mainly of *Sphagnum*), with notable layers of minerogenic material, until 900 yr BP, when it becomes a coarse, undecomposed peat, with similar gravel and sand layers distributed throughout the time period. Following on from this shift, there is one period of enhanced sedimentation rate, between 810 and 690 yr BP, where it rises to 0.22 cm/yr, as opposed to the normal rate of 0.1–0.15 cm/yr throughout the record.

The remainder of the core is characterised by several MM rises, a sign of near-constant minerogenic influx. The MM record indicates values between 60 and 90%, with no return to normal peat deposition, suggesting frequent mass wasting events, regular inundation of the bog, or a combination of both, between 350 and 100 yr BP. A very short period of low MM, fresh undecomposed peat is observed in the final 50 years, indicating a cessation of the major minerogenic deposition typical of much of the record.

4.2. Pollen

The pollen assemblages have been grouped into Local Pollen Assemblage Zones (LPAZ), constraining major shifts in the local vegetation type (Figs. 3–5).

LPAZ 1: 7500–5800 yr BP

This pollen zone is indicative of a mixed forest dominated by the upland *Picea*, with typical percentages between 40 and 50%, and a collection of more foothill representative taxa including *Alnus*, *Corylus* and *Quercus* typically making up 10% of the assemblage. *Carpinus* and *Ulmus* percentages are the highest of the core, with *Carpinus* making up as much as 15%, while *Ulmus* is typically 5% of the assemblage (Fig. 4). In addition to the tree taxa, high values for monolete spores are found throughout LPAZ 1, the only major component seen that is not arboreal.

LPAZ 2: 5800–4300 yr BP

This zone is characterised by a dominance of subalpine forest taxa: *Picea* (up to 60%) and *Pinus*. There are lower abundances of foothill forest taxa pollen, although *Carpinus* still makes up 10%, *Quercus* roughly 10% and *Fagus*, for the first time, becomes a clear component of the vegetation. The appearance of *Fagus* follows on from the disappearance of *Ulmus* at around 4500 yr BP. At the start of this zone *Corylus* drops to around 5% and never recovers to the values seen previously. In terms of non-arboreal taxa, an increase in Chenopodiaceae, and decrease in undifferentiated monolete spores may be observed.

LPAZ 3: 4300–3000 yr BP

Through this period, the coniferous pollen percentages begin to fall, with *Picea* dropping the most (to around 40% by the end of the zone), whilst *Fagus* (reaching 20% by the end of the zone), and *Quercus* increase. *Pinus*, which rises briefly early in the zone to 10%, drops back down to around 5% by the end. This occurs concurrently with an increase in *Sphagnum* and fungi spores, and a decrease in *Alnus* and undifferentiated monolete spores. *Carpinus* still makes up a fairly major component, with values of 10% common, until the end of the zone, where it disappears. *Dryopteris*, previously present in low percentages, disappears, and Poaceae, previously only observed in small pollen abundances, rises to 10%, by the end of the zone.

LPAZ 4: 3000–1900 yr BP

This period is characterised by another major decrease in *Picea* from 40% to 30% between 2750 and 2500 yr BP, a value around which it hovers for the remainder of the record. This decline is offset by an increase in *Alnus*, up to over 20% and *Quercus* (circa15%). At the same time, *Fagus* declines below 20%, and *Carpinus* disappears. Herb taxa, particularly Poaceae, Apiaceae, *Artemisia* and Asteraceae appear. The pollen data for this section indicate further shifts away from conifer-dominated forests. *Alnus* and *Fagus* (both as high as 30%) are dominant throughout the period, with *Picea* making up the remainder, indicative of more local deciduous forest.

LPAZ 5: 1900–950 yr BP

The pollen record from this period is indicative of a deciduous forest but with a contribution from coniferous taxa. *Picea* (30%) and *Pinus* (5%) remain low, with *Alnus* (25%), and in particular *Fagus* (30% rising to 40%) make up the majority of the assemblage. The herb and shrub taxa are at the highest levels yet seen, indicating the least dense forest, alongside the initial appearance of a number of taxa related to anthropogenic activity. Indeed, throughout this period, the abundances of *Plantago* and Poaceae are high, and the first occurrences of *Papaver* (1100 yr BP) and *Cerealia* (1450 yr BP), unequivocally agriculture-related taxa, are seen. The concurrent increases in Apiaceae Asteraceae, Chenopodiaceae, Ericaceae, and all further indicate lower forest cover.

LPAZ 6: 950 yr BP to 0 yr BP

Within this zone the vegetation remains dominated by *Fagus*, composing up to 40% of the assemblage, prior to a major drop at the end of the zone, at the transition into LPAZ 7. *Alnus*, *Quercus* and *Picea* make up the remainder of the arboreal taxa.

LPAZ 7: 0 yr BP to Present

The period is notable for the sharp increase in Poaceae (up to 15% by the end of the zone) and Chenopodiaceae percentages, recording the highest values of these taxa, and an extremely large spike in Cyperaceae, rising to 15%. Additionally, *Sphagnum* rises to 5%. The impact of this change may also be seen in the pollen concentrations which are at their lowest throughout the core (Fig. 4). Another notable feature is the highest abundances of *Cerealia*, *Papaver* and *Plantago*, all related directly or indirectly to agriculture or pasturing.

4.3. Charcoal

Between 7500 and 4550 yr BP, the microcharcoal record is characterised by high, but fluctuating values, between 100 and

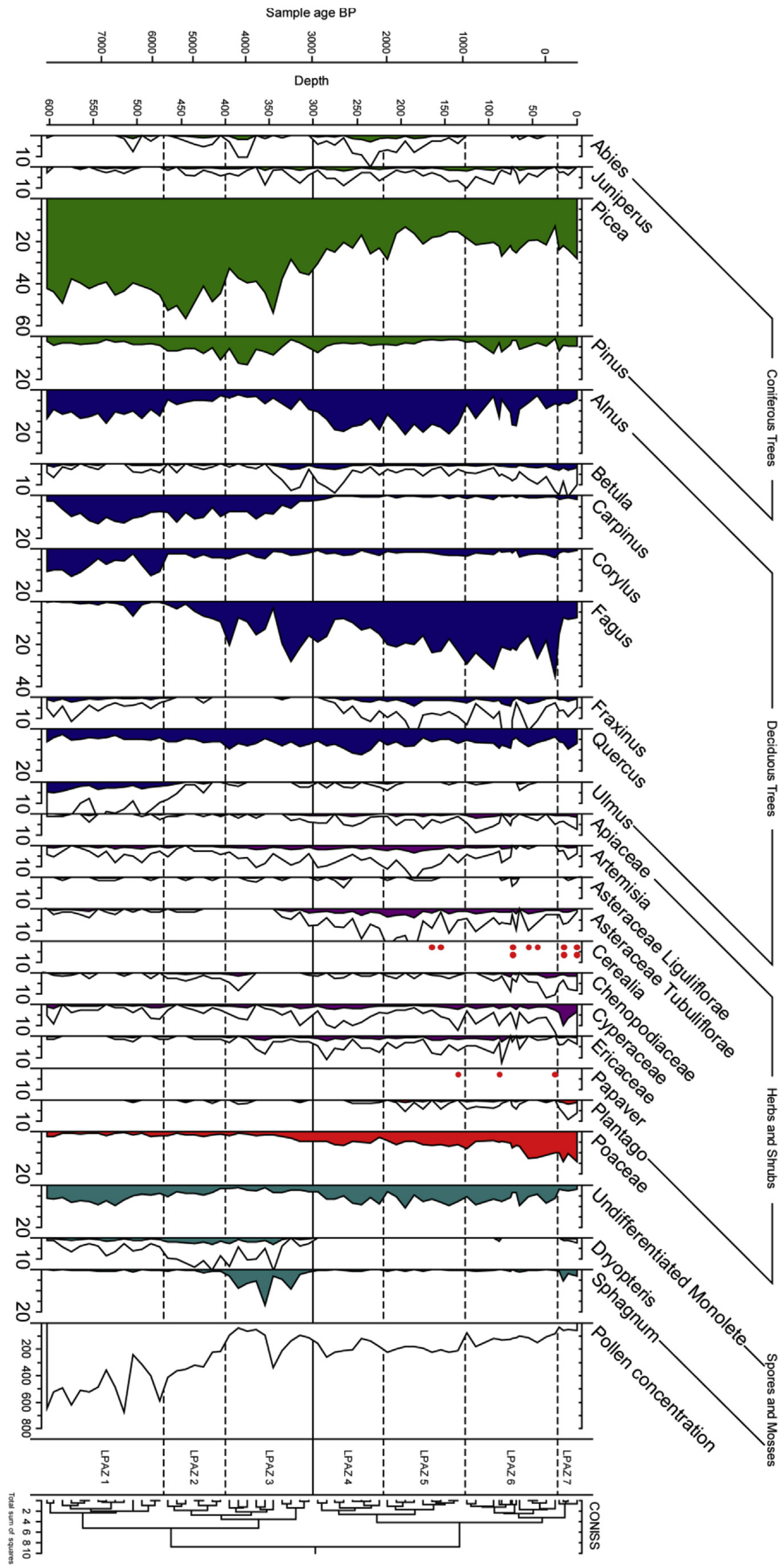

Fig. 4. Simplified pollen diagram showing percentages of selected taxa from Sureanu bog. Non-patterned, unshaded area represents five times exaggeration of percentages. Red circles are representative of one pollen grain. Percentages of pollen and spores were calculated based on the total pollen sum, excluding unidentified pollen/spores. Taxa used to reconstruct human impact have been highlighted in red. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

450 cm^2/cm^3 before an extended period of low (<50 cm^2/cm^3) values until 2740 yr BP (Fig. 3B). After this point, values remain high, ranging between 100 and 300 cm^2/g until a recent drop to below 50 cm^2/cm^3 in the final 50 years of the record.

4.4. Geochemistry

For the first 2500 years of the record (Fig. 3), the lithogenic elements indicate elevated concentrations (Rb: 100 ppm, Sr: 110 ppm, Sc: 5–10 ppm), indicative of a lacustrine system, with Rb values ~100 ppm having been observed previously in Alpine lakes (e.g. Koinig et al., 2003). These are significantly lower for the subsequent period, between 5050 and 2650 yr BP (Rb: 25 ppm, Sr: 50 ppm, Sc: 3.5 ppm). After 2650 yr BP, the values increase indicating higher mineral input, potentially as a result of enhanced regional erosion. Additionally, in this zone some trends in the Rb/Sr ratio appear visually coupled with the organic matter record (See Fig. 6).

The concentrations for all elements remain generally high for the section 2650 yr BP to present, with values of Rb: 200 ppm, Sr: 200 ppm, Sc: 10 ppm common. Short periods of lower values may be seen between 1950 and 1650 yr BP before high but fluctuating values between 1650 and 600 yr BP (Fig. 3). This is followed by periods of lower values with one peak, at 750–650 yr BP. The values return to the low concentrations observed earlier towards the top of the record, but not before a short period of enrichment between 200 and 50 yr BP with Rb (100–200 ppm), Sr (100–150 ppm) and Sc (5–10 ppm) showing short-term increases.

4.5. Grain size

Grain size analysis (Figs. 5 and 6), performed on the last 2500 years of the record, indicates two main types of grain size profile. First are periods of low d50 (average grain size 30–50 μm) which generally occur during periods of high organic matter (<35% MM). Within periods of high MM (>35%) d50 is generally higher, with values between 60 and 80 μm common, and reaching as high as 130 μm. Such high grain sizes are most prevalent between 2500 and 2000 yr BP (Fig. 5).

5. Discussion

5.1. Depositional events record

Sequential periods of peat growth (generally <20% minerogenic material) are interrupted by periodic, abrupt (in terms of boundaries) and short-term episodes of much higher MM values (generally <60%, but reaching as low as 10%, Fig. 3F). Due to the low values reached without a clear change in the peat material (still dominated by *Sphagnum* remains), these layers may be associated with the input of minerogenic debris from a proximal source, and not just direct atmospheric deposition of fine-grained particulates/dust. If it were simply atmospheric dust deposited within a raised bog environment, the OM values would remain much higher (roughly around 90%; Shotyk, 2002).

The age model can be potentially complicated by these minerogenic layers. If these are indicative of rapid deposition through mass wasting then they may lead to an incorrect age model. However, the Sureanu pollen profile matches similar sites in the region (See SI 3) and the sedimentation rate is relatively constant throughout (0.05–0.15 cm/yr) suggesting that the minerogenic deposition has had little impact on the sedimentation rate for the past 2500 years. Furthermore, observations of the core indicate the minerogenic debris was generally made up of fine-grained particles, with only very occasional pebbles, and so it is reasonable to assume such events do not cause cessation of peat growth (See SI 4). To further evaluate the robustness of the age model we compare our data with other pollen records from similar sites, with emphasis on the timing of appearance and disappearance of key species (See SI 3). These approaches appears to indicate the Sureanu record is replicating the trends observed regionally and is therefore based on a well-constrained age model.

5.1.1. Interpretation of minerogenic event layers

To understand the underlying source of the depositional events, grain size analysis has been performed on the top 2500 years of the record (since this section contains the majority of minerogenic debris layers), the sediment from the rocky shore of Iezerul Sureanu Lake, and a soil sample from the surrounding forested slopes between the lake and the bog (Fig. 1). Peat intervals with high organic

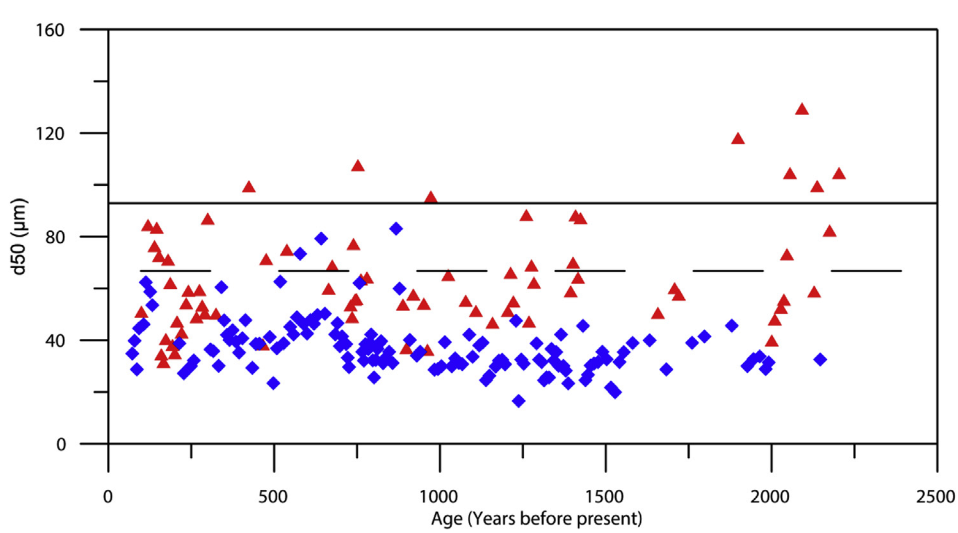

Fig. 5. Median grain size data (d50) for most recent 2500 years of Sureanu core. Samples taken from layers of minerogenic (<65% OM) are indicated by red triangles, whilst those from peat (>65% OM) are indicated by blue diamonds. Also indicated are the d50 values for both the local soil (solid line) and lake sediment (dashed line). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

matter values (>80%), show d50 (average particle size, in μm) values typically between 20 and 40 μm, (Fig. 5A). As the analytical method for determining grain size removes all organic material, and typically atmospheric dust particles are around 20 μm (Stuut and Prins, 2014), this can be considered to be the signature of normal atmospheric dust input.

Within minerogenic layers (as determined from the OM and the geochemical records) the grain size (compared to normal peat deposition) shifts, with d50 averaging around 40 μm and reaching as high as 130 μm (Fig. 5). This shift indicates the clear input a non-dust minerogenic fraction, with the site's location at the base of a cirque likely to be the cause. As the surrounding slopes are steep, and there are avalanche channels present, it is reasonable to assume the minerogenic debris present is related to slope activity.

It has been suggested (Nesje et al., 2007; Vasskog et al., 2011) that snow avalanches result in the deposition of coarse-grained sediment, as the snow pack breaks up and transports local rock as it moves downslope. When avalanches settle and the snow melts, these grains are deposited onto the substrate below (Nesje et al., 2007). Within Sureanu cirque, as the lake is regularly frozen during winter, avalanches would move over and the debris settle over the lake and bog. Additionally, mass wasting not related to snow action (i.e., landslides, rockfalls, flooding) would cause the same type of deposit as they break up the surrounding rocks into small particles as they flow down the slope (see Panek, 2015 for a review of dated landslides in sedimentary archives). Such activity is likely to be behind the shift toward larger particles during minerogenic layers. Deposits of this type would also result in the existence of large grains (>1 mm), as seen in the grain size distributions, core observations (clear pebbles may be seen within minerogenic layers), and in the presence of boulders in the lake and surrounding area.

Other depositional events appear to indicate the influence of the local soil being washed into the bog. The local soil has d50 values above the typical peat d50, but below that of the lake sediments (Fig. 5A). The simplest explanation for the presence of these deposits is the impact the hydrology of the nearby lake has on the site. When water levels in Iezerul Sureanu are high, the water overflows the bank on which the soil sample was taken, and washes into the bog, bringing with it sediment which has been entrained in the water, or picked up as the water flows over rock and soil.

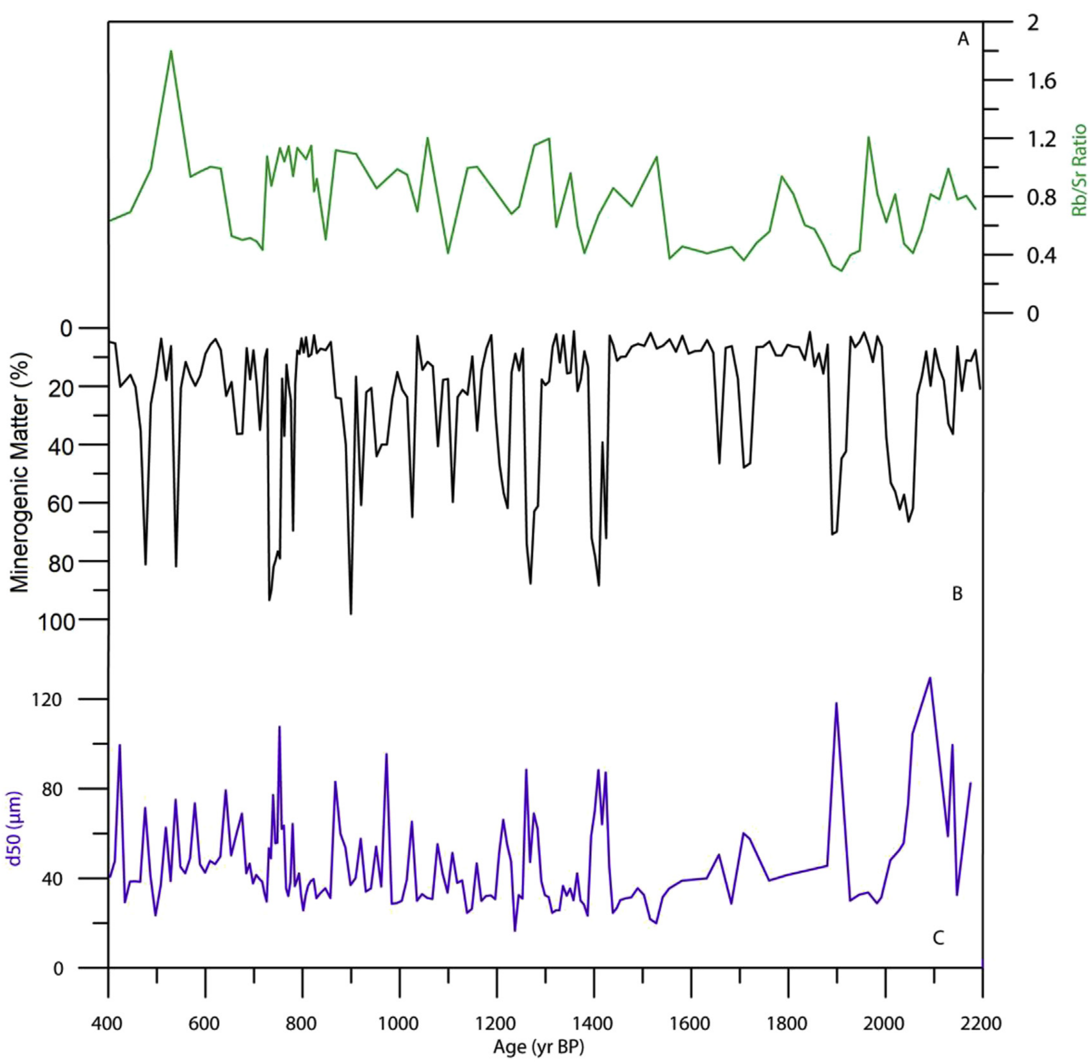

Fig. 6. Comparison of Rb/Sr ratio (A) to the organic matter record (B) and the grain size distribution (C) for the period 2200–400 yr BP, where tentative correlations may be made between the three methods for reconstructing grain-size variations.

5.1.2. Significance of the depositional record

For each of the methods of sediment delivery outlined above, water is a major control on their formation. For avalanches, the primary controller is high snowfall (Schweizer et al., 2003). The value of 30 cm of fresh snowfall is regularly used as an indicator for an increased avalanche risk (McClung and Schaerer, 2006). Studies in Iceland (Keylock, 2003) and the Pyrenees (Esteban et al., 2005; García-Sellés et al., 2010; López-Moreno et al., 2011) have shown that periods of high snowfall, and even large-scale atmospheric precipitation controls, such as the NAO, play a large role in the frequency of avalanches. Additionally, intense, short periods of rainfall are one of the major factors in landslide formation (Crozier, 2010; Popescu, 2002; Zaruba and Mencl, 1982) with a clear correlation between precipitation and landslide frequency. Further connections between NAO-controlled precipitation and landslide frequency have been made in the Azores (Marques et al., 2008), and Portugal (Trigo et al., 2005; Zêzere et al., 2008). As such, the minerogenic depositional event record may be used as an indirect proxy for periods of high precipitation affecting Sureanu (both bog and lake), and catchment area.

5.2. Investigating the potential of Rb/Sr ratio as a proxy for grain size

When investigating highly organic substrates such as peat, traditional grain size analyses are not easy to perform. Due to the high organic content, a large amount of sample is needed to extract a sufficient minerogenic fraction for analysis, as we have performed here. Furthermore, such approaches (removal via H_2O_2 or ashing) may alter the minerogenic debris, and so particle size measurements are not routinely performed (Kylander et al., 2016). Additionally, since there is no statistically reliable approach to quantify grains above 2 mm, a method which analyses the whole sediment signal may be useful. However, particle size analyses could be very useful for interpreting the provenance of both local (avalanche and mass wasting) and distal (dust) minerogenic inputs (Kylander et al., 2016; Vasskog et al., 2011).

The rubidium/strontium (Rb/Sr) ratio has been used in lake sediments from China to indicate weathering in the catchment area (Jin et al., 2001, 2006), and then to estimate weathering intensity in loess-palaeosol deposits (Jin et al., 2006). This work was developed based on the behaviour of the two elements during weathering. Rubidium commonly substitutes for K in mineral lattices, and Sr commonly replaces Ca, due to similar ionic radii (Kabata-Pendias, 2010; Vasskog et al., 2011). Minerals containing K are much more resistant than Ca-bearing minerals, resulting in an enrichment in weathering products of Ca (Boggs, 2013; Kabata-Pendias, 2010; Simmons, 1998). Consequently, Sr should be enriched in glacigenic, and weathered material (Vasskog et al., 2011). In Norway, the proxy has been used to establish a preliminary record of mass wasting events from the slopes surrounding a lake (Vasskog et al., 2011).

The use of Rb/Sr ratio has been further refined, and it appears that the ratio also changes as a function of grain size (Kylander et al., 2011), or more precisely, correlates with the lack of clay in the layers of debris (Chawchai et al., 2015; Vasskog et al., 2012). This theory rests on the K-bearing minerals being derived primarily from finely crushed phyllosilicates, whilst Ca-bearing minerals are concentrated in coarser, more erosion-resistant grains, with plagioclase a common constituent. This results in low Rb/Sr ratios associated with large grain sizes.

The periods between 7500 and 2950 yr BP, and from 200 yr BP to present appear to show no direct coupling between the Rb/Sr and the minerogenic record, other than the reduction in Rb/Sr at 5000 yr BP mirroring the decrease in MM. This appears to indicate another major control on the distribution of these elements within the core during these times (Figs. 4 and 6). The results appear to indicate the issues of applying a proxy previously utilised (Vasskog et al., 2011) where the influence of organic matter is negligible, to an organic-rich substrate.

Within the period 7500-2950 yr BP the Rb/Sr ratio is uncoupled from the MM, related to the lacustrine nature of the sediment, with high values to be expected. During this time the clayey lacustrine sediment would be associated with fine grain sizes, and may explain the ratio shift. This is corroborated by the LOI record (Fig. 3), which indicates a shift towards clay-rich peat, and lower organic contents. The period of very low ratios between 300 and 375 cm appears to be due to a natural shift into a gyttja-like sediment, at this time, which contains larger grain sizes (Fig. 3).

In the upper 50 cm, the high Rb/Sr values may reflect the position of the water table, and the shift to more stable values could be indicative of the contact between the aerobic (acrotelm) and anaeorobic (catotelm) peat, with the shift from active to inactive peat controlling the elemental distribution. The decomposition process and transition from live material to inactive causes the increased migration (out of the layer of interest) of Rb relative to Sr (Tyler, 2004, 2005). Additionally, due to the ease with which Rb and Sr may be taken up by plants (Kabata-Pendias, 2010), it can be assumed that both elements will be affected by migration. The discrepancy could be a result of Rb being taken up more readily than Sr, and easily cycled by organic compounds due to its lower atomic weight and electronegativity (Tyler, 2005), and substitution for K in upper peat layers. Rb is particularly likely to be re-absorbed by the upper organic layers when the system is acidic (Folkeson et al., 1990), as it is the case of Sureanu bog. It has been demonstrated that acidity can exacerbate the uptake by causing K^+ leaching losses, resulting in replacement by Rb^+ where available (Nyholm and Tyler, 2000), especially when mediated by intense fungal activity (Vinichuk et al., 2010), which would be concentrated in the upper peat layers.

Between 2950 and 200 yr BP some similarities may be observed between Rb/Sr ratios and the MM content, with most of the major rises in the MM values corresponding with a lower Rb/Sr ratio (Fig. 6). Typically, the periods of highly organic, undisturbed peat produce Rb/Sr values around 1, with the ratio dropping to as low as 0.4 in the debris events. Therefore, for at least this part of the record, the Rb/Sr ratio correlates with the MM record, or weathering products associated with mass wasting. To corroborate this, the Rb/Sr ratio of both the local lake deposits (0.55) and bedrock (0.35, calculated from values given by Rudnick and Gao (2013) from the average continental crust) have a value close to the lower, debris event related values. This could indicate either directly weathered bedrock or lake sediment entering the bog as a result of the mass wasting processes to be the source of the low Rb/Sr ratios.

However, quantitative comparisons indicate only a weak correlation, with an r-value of 0.08 throughout the record, and 0.1 for the past 2500 years, although the p-values (<0.001 for both, $n = 75$) indicates significance. Further, wavelet coherence and cross wavelet analysis have been performed, indicating little clear correlation. This is to be expected, however, since Rb/Sr is perceived to be related to grain size fluctuations, and different types of minerogenic input (e.g. avalanches, flooding, see Fig. 5) each produce different Rb/Sr ratios, potentially complicating the geochemical signal, which is then compared to an ‘averaged’ minerogenic signal (the MM values). This is further evidenced by direct comparison of Rb/Sr with grain size, where no correlation ($r^2 = 0.042$) is observed, likely due to the inability of the particle size analyser to provide any data on the very large grains (as its upper limit is 2 mm), whereas the geochemical approach analyses the entire signal. As a result of the lack of statistical correlation, our recommendation is that utilisation of Rb/Sr alone is insufficient for reconstruction of grain size fluctuations in organic-rich sediment. However, the apparent visual similarities indicate some form of relationship which needs further validation.

5.3. Palaeoenvironmental reconstruction from Sureanu bog

The mid Holocene (7500–4500 yr BP) is characterised by the absence of major detrital events and relatively little variability in the minerogenic matter record. Vegetation throughout this period consists primarily of conifers, with some input of pollen from lower altitude taxa, a combination which is indicative of the natural vegetation at this altitude in this region (Feurdean et al., 2010). Additionally, the undifferentiated monolete spore content is high, related to mosses and ferns, and potentially an indication of wet conditions, as also inferred at a nearby site for this period (Buczkó et al., 2013). These wet conditions are reflected in the deposition of gyttja at the site, and a shallow lake environment throughout this period.

After 5500 yr BP, the rise in *Picea* and *Pinus* pollen indicates expansion of conifer forests, potentially pointing to a cooler climate. This interpretation is supported by cool conditions having been observed in a chironomid-inferred temperature record from Retezat Mountains nearby (Tóth et al., 2015), and in the Maramures Mountains in NW Romania (Fărcaş et al., 2013). From about 4600 yr BP, there is a stepwise change away from low-organic gyttja and the organic content increases gradually until ~3500 yr BP after which the MM remains low. Occasional low MM events present between 4500 and 3500 yr BP signal the initial development of peat bog, but are interrupted by transitions back to gyttja.

At 3500 yr BP, there is a shift to coarse, detritus-rich peat, reflected in the decrease in lithogenic element (Rb, Sr) concentrations, likely reflecting the initiation of the gradual process of raised bog formation. This is corroborated by the development of the site from a much wetter, gyttja-depositing environment to a peat bog. This is likely determined by a reduction in the water being supplied to the site and the natural infilling of the pre-existing lacustrine basin. Limited input of water would mean overall wetness is reduced, and would allow for peat growth, rather than peaty gyttja, which is generally formed in waterlogged conditions. This transition to ombrotrophy and raised bog formation results in subsequent sedimentation occurring above the water table, and accounts for the reduction in lithogenic elements, and increase in organic matter (Charman, 2002).

The change in sediment type is accompanied by a marked decrease in *Picea* values and an increased abundance of the mid-altitude taxa *Fagus.* This is a significant shift in vegetation composition, but represents a change seen in other regional studies (Feurdean et al., 2011 and references therein). It is possible that the decline in *Picea* was in response to regionally cooler summers and wetter conditions (Onac et al., 2002; Schnitchen et al., 2006; Feurdean and Willis, 2008; Magyari et al., 2009; Tóth et al., 2015; Drăguşin et al., 2014), superimposed on a slightly higher winter insolation relative to the early-mid-Holocene (Berger and Loutre, 1991). *Fagus* is much more sensitive to colder winters than *Picea* (Feurdean et al., 2011), and increased winter insolation and associated warmer winters would have allowed its spread in the region. Its ability to thrive in shade during its juvenile stage allows for it to out-compete *Picea* during periods of climatic warming (Feurdean et al., 2011).

Prominent and abrupt rises in MM values characterised the peat deposition after 3500 yr BP (Fig. 3). The values never rise above 50%, and always return to a baseline value of around 10% soon after, indicating that the bog environment went quickly back to normal peat formation, before the next minerogenic event occurred. If the mechanism for these isolated events is increased precipitation and subsequent inundation of the bog during Iezerul Sureanu Lake highstands, it would parallel the signal seen within local (Buczkó et al., 2013) and regional records (Cristea et al., 2013; Drăguşin et al., 2014; Gałka et al., 2016; Lotter and Birks, 2003; Magyari et al., 2009; Onac et al., 2015; Schnitchen et al., 2006). Alongside the appearance of detrital events, a rise in hydrophilic taxa (*Sphagnum* in particular) is observed as a reaction to this regional precipitation increase. Additionally, the charcoal record indicates a period of extremely low fire activity between 4000 and 2500 yr BP, which could also be the result of increased precipitation that limited biomass ignition. Prior to this, regional fie activity was higher, but with large fluctuations in amount of charcoal deposited (Fig. 3) and so onset of this low fire regime appears to be controlled by regional precipitation.

The period of wetness implied by the low fire regime is followed in the pollen record by an increase in *Alnus* and *Quercus* and a further decrease in the previously dominant *Picea* between 2500 and 2000 yr BP, indicating a decrease in conifer forests, as documented regionally (e.g. Finsinger et al., 2016), in response to local cooling (Tóth et al., 2015). The major shift away from a boreal forest to a more deciduous one, and in particular, the replacement of *Picea* with *Fagus* is a trend recorded in various other local and regional vegetation studies (Tanţău et al., 2006, 2011; Feurdean, 2005; Feurdean et al., 2009, 2011, 2015; Magyari et al., 2009). Alongside this, the disappearance of *Dryopteris* - ferns typically found in the understory of dense forest appears to indicate a further regional decline in forest cover. Alternatively, this may be related to a change in preservation conditions, resulting in the identification of *Dryopteris* spores as undifferentiated monolete spores (See Fig. 4).

After 2500 yr BP there is an increase in lithogenic elements, indicating higher mineral input, potentially as a result of enhanced local erosion, linked to precipitation-controlled weathering, a process that reduced tree coverage would exaggerate. In the Retezat Mountains, a temperature rise is observed at 2200 yr BP (Tóth et al., 2015), and it is therefore possible this change in vegetation could reflect the impact of the so-called Roman climatic optimum, a period of warm and dry conditions in the region (Büntgen et al., 2011a,b). In addition to natural changes, warmer temperatures would allow increasing human use of uplands, for pasturing, thereby reducing forest cover, and increasing erosion. Furthermore the charcoal increase at 2500 yr BP may be linked to such increased human activity on the high mountain environments of Sureanu (Finsinger et al., 2016).

After 2500 yr BP, the minerogenic depositional events become more frequent and pronounced, becoming particularly regular after 1500 yr BP (Fig. 3). Much of the early part of this period corresponds to the Medieval Climate Anomaly (MCA), with generally warm conditions seen in the Northern Hemisphere (Christiansen and Ljungqvist, 2012). Locally, the MCA is characterised by an increase in wetness (Feurdean et al., 2015). However, it is not reflected in the July temperature reconstruction from tree rings as presented by Popa and Kern (2009). At Sureanu, the interval between 1150 and 850 yr BP is characterised by periods of large-scale mass wasting, with a large number of low OM events. As discussed earlier, a precipitation increase would increase weathering rates, and it is possible events through this period are associated with major rainfall leading to large-scale slope erosion, and flooding of the lake adjacent to Sureanu bog. This may be equivalent to an episode of intensified erosion signal seen within Sf Ana Lake in the Eastern Carpathians (Magyari et al., 2009), interpreted as a reflection of deforested slopes in the area.

A short period of normal peat growth follows, before the most pronounced stretch of minerogenic deposition between 350 and 100 yr BP. The initiation of this period of intense minerogenic deposition is closely correlated with the onset of the LIA (Mann et al., 2009) which is generally associated with cooler climate (Bradley and Jonest, 1993; McGregor et al., 2015). Regional palaeoclimate reconstructions indicate the initiation of this period at around 300 yr BP (Popa and Kern, 2009; Feurdean et al., 2015) (Fig. 3). The MM signal rises at almost exactly the same time, suggesting that the increased deposition of clastic material within the bog is associated with regional climate forcing. Increased erosion during the LIA has been observed in a number of places, from the Alps (Wilhelm et al., 2012, 2013; Arnaud et al., 2012) to mardels in Luxembourg (Slotboom and van Mourik, 2015), but not documented in the Carpathians before. The signal seen here therefore may be attributed to the cooler and wetter climate causing increased rainfall in summer and snow avalanches events in winter period. Other local studies do not define well the extent to which the LIA had an impact on the area. Our pollen data for this time also indicates no clear shift in vegetation, echoing other vegetation and other palynomorph records, which generally indicate no specific signal (Tanţău et al., 2011; Buczkó et al., 2013; Tóth et al., 2015). Additionally, available speleothem isotopic proxies have too low a resolution to discern LIA-related changes (Onac et al., 2002; Drăguşin et al., 2014). A clear drying is indicated via testate amoebae (Schnitchen et al., 2006), and diatoms (Buczkó et al., 2013) but this drying does not appear to be specific to the LIA as in both cases it does not cease with the end of the period. Therefore, this may be the first clear indication of the impact the LIA had on the Sureanu bog record: a period of intense minerogenic deposition, from runoff-sourced erosion and lake flooding denoting increased precipitation rates.

5.4. Human impact

Up until 3650 yr BP, human impact at the site appears minimal, with only sporadic *Plantago* (from 6200 yr BP onwards) and Poaceae (present from the base of the core) pollen indicating anthropogenic influence via deforestation. Indeed, it is plausible that humans had begun to clear forests for agriculture more regionally (Schumacher et al., 2016), but not necessarily in the local environment, although the alpine environments in the Carpathians are to the current day heavily exploited for pasturing. Additionally, as the climate during this period appears fairly humid, high charcoal values (Fig. 3) could be an indication of man-driven, regional biomass burning. Human impact has been observed via the appearance of grass and crop taxa as far back as 8000 yr BP in the Northern Carpathians (Feurdean, 2005; Fărcaş et al., 2013) and 7500 yr BP in the nearby Transylvanian Depression (Tanţău et al., 2006), so it is possible these are the first hints at pasturing activities proximal to our site. However, it is unlikely humans had much of an impact through this period at the Sureanu site.

At 4200 yr BP, a large decrease in pollen concentration occurring at the same time as the first drop in *Picea*, corresponds to the onset of the Middle Bronze Age in Romania (Bailey, 2000). This is somewhat earlier than most regional studies place the first major impact of humans in the region, with 3200 yr BP being typical in the Romanian Carpathians (Tanţău et al., 2003; Feurdean et al., 2010; Tanţău et al., 2011). Due to the location of Sureanu bog near the upper edge of the treeline, it is possible that this site is more sensitive in recording traces of early small-scale deforestation than these other sites. At this time, the local Wittenberg and Verbicioara cultures are both known to have developed copper extraction and smelting (Gimbutas, 1965; Gogâltan, 1995), which would have required exploitation of other natural resources, including timber. This anthropogenic disturbance occurs concurrently with the climatic forcing previously mentioned.

The second decrease in *Picea* after 2900 yr BP (Fig. 4), is not seen in many other regional vegetation reconstructions, with the *Picea* percentages remaining fairly stable after an initial drop at around 3000–4000 yr BP (Feurdean et al., 2011; Tanţău et al., 2011). The use of *Picea* timber for building, and of denser wood like *Carpinus* and *Fagus* as fuel for metal smelting in the area (with the northern valleys draining Sureanu Mountains particularly rich in gold placers and tin) may explain these shifts, and potentially the disappearance of *Carpinus* and the drop of *Fagus* at this time. This initial drop is followed by further reductions in *Picea*, (between 2900 and 2000 yr BP). It is possible this is related to climatic change, but it correlates well with the advance of Late Bronze Age and Dacian people, and the growth of their economy, based mainly on the mineral resources of the Carpathians, particularly gold, silver and iron (Mountain, 1998; Anthony and Chi, 2009) and also extensive agriculture. A strong human influence is expected at our site as the Dacian capital, Sarmizegetusa Regia (a well-known centre for metal smelting in Antiquity), was established 15 km to the west around 2200 yr BP at 1030 m a.s.l (Oltean, 2007). This may also be inferred from the charcoal record, indicating a large shift at around 2900 yr BP from extremely low values to much higher levels which characterise most of the last 2000 years. The relatively stable climate over this period (Onac et al., 2002; Schnitchen et al., 2006; Buczkó et al., 2013) provide additional support for the anthropogenic influence on the observed changes in vegetation.

Soon after, the first large perturbations in the MM record occur, dated between 2200 and 1800 yr BP. This was a period of great upheaval in the history of the region, with the establishment of the Dacian Kingdom, with the nearby Sarmizegetusa Regia as its capital and part of a network of fortresses and high-altitude settlements encompassing the Sureanu Mountains on all sides, followed by the Trajan wars with Rome (AD101–106), and subsequent incorporation of Dacia into the Roman Empire (Gudea, 1979; Bailey, 2000). These large fluctuations in the MM record could be an indication of the local impact war had on the region, presumably with large-scale tree felling for weaponry and defences common (Hughes and Thirgood, 1982). Additionally, the charcoal record indicates the prevalence of fire events through this period; it is likely that many of these fire events were not natural. The remains of a Roman military castrum located on Varful lui Patru at 2100 m altitude, in the vicinity of the Sureanu bog, further supports evidence for a strong human impact at the time, even at such high altitudes. The mass wasting events do not cease until the final 100 years of the record, so it is likely the local environment never recovered fully from the deforestation and impact humans inflicted upon the site area starting in the Antiquity period.

After the collapse of Roman Dacia, in 271 CE, the charcoal content decreases, suggesting a peak at around 200 CE is potentially an indication of a shift of the economy of the region from primarily mining and metal production towards agriculture (Poulter, 2007), which persisted throughout the Middle Ages. These pastoral communities are unlikely to have produced the same scale of clear-felling as major Roman or Dacian activity, and may explain the reduction in fire events throughout this period, and the relatively stable vegetation assemblage of the last 1800 years. The appearance of the first unequivocal agriculture-related *Cerealia* at 1400 yr BP confirms this shift toward widespread farming.

The pollen record for the final 500 years indicates the clearest human impact, with increased *Cerealia*, *Plantago*, Poaceae and other herb taxa (Fig. 4). This increase is particularly noticeable in the last 200 years, an indication of major forest clearance in the region, as noticed in many other regional studies (e.g. Feurdean et al., 2008a,b; Tanţău et al., 2011). The most recent section of the core (50 yr BP to present) indicates no clear minerogenic events (Fig. 3). The increase in temperatures seen over the last five decades has had a negative impact on the amount of snow in the region (Birsan and Dumitrescu, 2014) with an overall decreasing precipitation trend over the period. It is likely the reduced snowfall has led to a decrease in the number of avalanches and sudden snow-melt events, as, despite other causes, the primary controlling factor on mass wasting is still the amount of snow (Esteban et al., 2005). The warmer summers of this period (Popa and Kern, 2009) appear to have had no effect on the number of summer rainfall events. Additionally, regional reconstructions indicate clear drying throughout this time period (Schnitchen et al., 2006), explaining the cessation of mass wasting processes. The correlation of mass wasting ceasing and reduced winter snowfall indicates the likelihood that the main driving force behind the mass wasting processes around Sureanu bog is snow availability.

5.5. Comparison of Sureanu with other records of palaeohydrology and flooding

As it appears that the primary controller of the minerogenic deposition in this environment is precipitation-related mass wasting, with occasional lake flooding events, we compare the flooding signal as reconstructed using the Sureanu record with other palaeohydrological and flooding reconstructions (Fig. 7). As there are very few (Haliuc et al., 2017) such reconstructions from south-eastern Europe, we compare to a selection of central European records.

Within Sureanu bog flood layers appear to be rare prior to around 2000 yr BP. This sporadic flooding is similar to that which is observed in the Ammersee (Germany), the Mondsee (Germany) and Polish rivers at a similar time, with small, intermittent flood layers common in all sites (Czymzik et al., 2013; Starkel et al., 2006; Swierczynski et al., 2013). Clear intensification of the number of flood events is present from 2800 yr BP onwards in the Ammersee, 2000 yr BP in Poland and after 1500 yr BP in the Mondsee. Within Sureanu, this intensification may be seen at either 2000 yr BP (Figs. 3 and 7), when the first large minerogenic input occurs, or at 1350 yr BP, when the onset of nearly constant deposition minerogenic is clearly documented. The central European flood records both ascribe the intensification to human activity and the effect deforestation had on the source area. The Sureanu pollen record shows that the major drop in tree taxa, and inferred deforestation earlier, roughly 3000 yr BP (Figs. 3–4), and so it is sensible to assume the onset of minerogenic depositional events has presumably been mediated mainly by climatic factors.

After 2000 yr BP specific periods of enhanced flooding may be correlated to other records, with flood activity in the southern Alps appearing to show the same period of intense flooding 1200–900 yr BP, and short term fluctuations thereafter (Arnaud et al., 2012; Wirth et al., 2013). In addition, a clear correlation between minerogenic deposition in Sureanu and high flooding in the Alps may be seen between 500 and 50 yr BP, during the Little Ice Age. The correlation here is very good, with all records showing the initial intensification at 500 yr BP, a short period of less intense flooding, then a further increase before a drop at 50 yr BP (Fig. 7). Clearly the climatic controls between the two areas during the LIA are rather similar.

The flood records of the Northern and Southern Alps are interpreted by Wirth et al. (2013) to be indicative of fluctuations in the NAO. Due to the location of Sureanu, at the far eastern edge of Atlantic-influenced area, small shifts in the strength of the NAO should be also detectable in our bog record. Indeed, it appears many periods of intense flooding do correlate to periods of weakened NAO (Fig. 7). When the NAO is weaker, Mediterranean air masses become more dominant, with extreme summer rainfall in the Carpathians as a result of low pressure systems forming to the south of the site (Parajka et al., 2010). In Romania, a positive (negative) NAO index is associated with negative (positive) precipitation anomalies (Bojariu and Paliu, 2001; see Fig. 1C) and reductions in snowfall (Birsan and Dumitrescu, 2014).

Within the Sureanu record, and particularly the past 1500 years, flooding periods fluctuate in time with relatively small decreases in the NAO intensity. This is indicative of the sensitivity of the area to climatic fluctuations, in accordance with model predictions of sensitivity in this location and altitude (Bojariu and Giorgi, 2005) and the first demonstration of such a connection in this area over the late Holocene. Such an impact has been seen in the Southern Balkans, with NAO-controlled palaeoenvironment fluctuations observed in Lake Butrint (Morellón et al., 2016), but not in the northern section, like the Carpathians. It is clear, however, that not all enhanced flooding periods may be attributable solely to NAO fluctuations (e.g. 1100–1050 and 900–850 yr BP). These are likely to be related to the interplay between the NAO and other major climatic systems in the area, and indicate that unlike the Alps, the NAO is not the only major forcing factor at play in the Carpathian – Lower Danube area, and that its influence is periodically weakened; indeed it appears the eastern edge of dominant NAO influence is found at around 30°E (Krichak et al., 2002).

Signal from more than one major Sea Level Pressure (SLP) pattern is to be expected, with sites to the south east, including Lake Nar (Turkey) (Jones et al., 2006), Soreq (Bar-Matthews et al., 1997) and the Dead Sea (Migowski et al., 2006) showing no NAO-related connectivity, whilst sites further west show clear correlations (e.g. Wirth et al., 2013). Our record therefore indicates the decreasing impact of the NAO on climate as one moves from Western Europe east through to Eurasia. Furthermore, it provides a long time series showing evidence of the east-west climate see-saw in the Mediterranean (Magny et al., 2013; Roberts et al., 2012), with the reducing influence of the NAO being one of the major drivers behind it.

6. Conclusions

Using pollen alongside sedimentological and geochemical methods from a peatbog in the Southern Carpathians a regional record of the depositional environment and palaeoclimate has been produced.

We find that:

1. Both natural climatic fluctuations and human impacts are clear in the vegetation record of the site. Between 7500 and 4500 yr BP, there was a slow shift from relatively warm mixed forest towards cooler, conifer-dominated forest after 5000 yr BP, relating to natural increases in warmth and humidity in the region. From 4500 yr BP, the impact of human activity may be seen, with decreasing forest cover, and evidence for agricultural (mainly pasturing) activities in the high-mountain environment of Sureanu Mountains (from 3300 yr BP onward), alongside an inferred warming evidenced by the increase in deciduous taxa. This correlates with the onset of major agriculture in the region, and develops in time with shifts in local economy and development.
2. We present a record of minerogenic deposition likely forced by changes in hydroclimate, the first of its kind in this region. Sources of debris have been identified using grain size analysis, indicating input from periods of lake highstands, but with precipitation-related mass wasting being the main control. In addition, the Rb/Sr ratio has been utilised for the first time to determine depositional events within an organic-rich core, although the methods need further validation.
3. Particularly intense minerogenic deposition is observed during the Medieval Warm Period and the Little Ice Age, the first such indication of the effect these periods had in

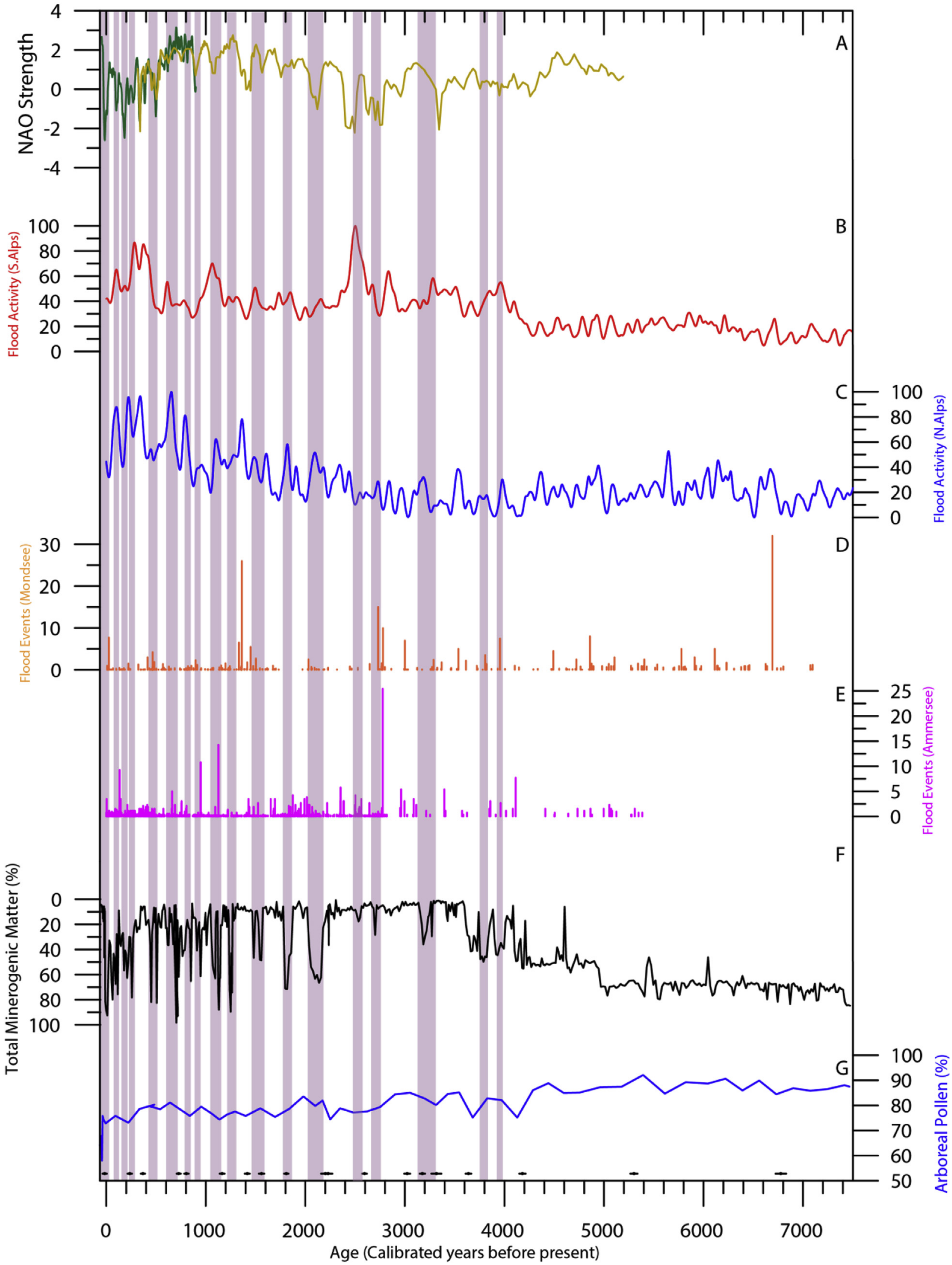

Fig. 7. Comparison of Sureanu minerogenic deposition record to Mid-Late Holocene climate forcing and records of palaeoflooding. Periods high minerogenic deposition are highlighted in purple, indicating correlation with other flooding records and the NAO. A) NAO Index as reconstructed by Trouet et al. (2009) in green and Olsen et al. (2012) in orange. Flood activity in the Southern (B) and Northern (C) Alps from Wirth et al. (2013) D: Flood events as indicated by Mondsee sediments (Swierczynski et al., 2013) E: Flood events as indicated by Ammersee sediments (Czymzik et al., 2013). F) Sureanu organic matter record. G: Arboreal pollen (AP) percentages from Sureanu; radiocarbon dates and uncertainties are indicated at the base of the graph. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

palaeoenvironmental records from the Carpathians. Warm and dry conditions over the last 50 years have led to a cessation of mass wasting in the local environment, indicating the reduction in precipitation and snow coverage is a major driver in the control of erosion in this region.

4. We infer a teleconnection with major atmospheric circulation patterns, as most fluctuations in flooding correlate to decreased NAO index values. This shows for the first time the long-term impact of the NAO in this region, which has previously only been predicted through modelling.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.quascirev.2017.04.029.

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