Microscopic charcoal and tar (CHAT) particles in peat: a 6500-year record of palaeo-fires in southern Sweden

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SUMMARY

Peat stratigraphies of eleven raised bogs in southern Sweden were investigated. Measurements included the occurrence of charcoal and various tar particles. Most of the particles found were microscopic, i.e. 5–100 µm in diameter. Two distinctly different groups of particles were distinguished: (A) charred fragments of plant tissue and (B) objects formed from tar, which were classified into five sub-groups on the basis of morphology. Both charcoal and tar are indicative of mire and forest fires. We suggest that it is possible to use the different groups of particles as fire regime indicators. Hence, the high frequency of charcoal and tar (CHAT) in the lower parts of the stratigraphies, i.e. in the lower strongly decomposed fen and carr peats that were formed before *ca.* 4000 cal ^{14}C BP, could be indicative of intense and frequent local fires. The decreasing abundance of CHAT and the lower relative share of Type A particles within the lower strongly decomposed *Sphagnum* peat *ca.* 4000–2500 cal ^{14}C BP signify a transition from local to regional fires. With a few exceptions, the uppermost weakly decomposed ombrotrophic peats formed after *ca*. 2500 cal ^{14}C BP, in which both charcoal and tar are rare, indicate a period of low fire frequency at both local and regional scales. There is no regional variation in the lower material, and it seems that wildfires were common phenomena throughout southern Sweden during the first few thousand years after peat formation began 6–8000 years ago. From a climatological point of view, the mass occurrence of CHAT in the lower parts of the profiles indicates a warm and dry Mid Holocene with frequent and widespread wildfires, and a moist and cool Late Holocene with more sporadic fires. Spectral analysis of the entire dataset shows significant periodicities of 610, 70, 30, 21, 17 and 14 years, the two most significant being 14 and 70 years.

KEY WORDS: charcoal and tar particle classification; fire frequency; late and middle Holocene

INTRODUCTION

Information on fire history is provided by two types of proxy data, namely fire scar records dated by dendrochronology and charcoal records from lake sediments and peat deposits. Whereas tree-ring information provides temporally precise short-term reconstructions of fire events, usually spanning the last 400 years or less (NOAA 2011), charcoal and tar (CHAT) particle records from sediments and peat can be used to reconstruct much longer fire histories, although with lower temporal and spatial precision (e.g. Olsson *et al*. 2010).

The occurrence of CHAT in peat may have different causes. In the first place, the production of such particles is related to the characteristics of vegetation and type of fire, including the supply and quality of fuel and the efficiency of combustion. The fires could have natural causes, such as volcanism (e.g. Scott & Jones 1996, Wilmshurst & McGlone 1996, Samaniego *et al*. 1998) or lightning (Scott & Jones 1996, McCafferty & Owen 1996); or human causes, i.e. slash-and-burn clearances and regular large scale burn-beatings.

The destiny of small particles produced by these fires is largely dependent on weather conditions. In the case of a strong and rapid fire lapse, the smallest particles carried up into the atmosphere at high vertical velocities by thermal uplift will simply fall to the ground locally if side winds are weak or absent. With stronger winds, however, these particles can be transported over vast distances before they are deposited under the influence of gravity or turbulent air flows (Whitlock & Larsen 2001). Deposition can be dry or wet, i.e. the microscopic carbonaceous particles can either drizzle down as dust or act as condensation nuclei in aggregates with sulphate (Posfai *et al.* 1997, Buseck & Posfai 1999) and fall with rain and snow.

The CHAT particles found in peat are either autochthonous particles formed by fires in the peatland, e.g. macroscopic or microscopic pieces of charred wood, herbs and mosses; or allochthonous microscopic particles brought in by wind and/or precipitation from more distant sources according to the mechanisms described above. Even large charred fragments can be uplifted in fire plumes and travel long distances (Whitlock & Larsen 2001). Whereas the macroscopic particles (> 125µm) generally provide evidence of local fires (Ohlson & Tryterud 2000, Gardner & Whitlock 2001), the microscopic particles (< 125µm) can provide clues to the locations of fires, and the data can be used to describe fire regimes at multiple temporal and spatial scales (Brussel 1988, Carcaillet *et al*. 2001). CHAT in peat can also be used to explain vegetation shifts in peatlands as well as in their surroundings, and dated charcoal horizons in peatlands may be used for dating such changes nearby (Alm *et al*. 1992). Whereas charcoal and tar particles in lake sediments do not necessarily reflect actual fire events because they could have been redeposited (Whitlock & Larsen 2001), CHAT in peat is expected to have been deposited at the time of peat formation at the specific stratigraphical level where the particles occur.

Charcoal accumulation may continue for a few years after a fire due to transportation and redeposition of allochthonous charcoal (Whitlock & Millspaugh 1996). This process tends to blur the exact age of a fire, even when charcoal particle ages are determined directly by radiocarbon dating. Additionally, the charcoal deposited may represent more than one fire within the area or fires from more than one year. As a result, fire episodes are referred to as one or more fires occurring within the time period of interest, rather than individual fires. The interval between fire episodes is generally longer in the sedimentological record than in the tree-ring record, suggesting human influence during the periods covered by the dendrochronological record (Pitkänen 2000).

Charcoal can be found not only in peats formed during the Holocene, but also as an indicator of wildfires in the palaeo-environments in which coal (Scott & Jones 1996) and lignite (Parish & Lambertson 1998) were formed. In the Holocene perspective, the occurrence of charcoal in peatlands shows large temporal variation (Wu-Hong 1992). In the early Holocene of southernmost South America, charcoal particles have been deposited intermittently since about 13 ka BP and in great abundance after 11 ka BP (Markgraf 1993). The mass occurrence of CHAT in the lower parts of many peatlands indicates a warm and dry mid Holocene with frequent and widespread wildfires, and a moist and cool late Holocene with more sporadic fires.

Charcoal and tar particles (CHAT) were examined during a study of mineral particles in peat from southern Sweden, which was carried out in order to investigate the deposition of extraterrestrial material on Earth. The black tar spherules which are common in the lower stratigraphical levels of these peatlands have a similar visual appearance to black magnetic spherules (Franzén 2006, Franzén & Cropp 2007), and can easily be mistaken for such without geochemical analysis. Initially, charcoal and tar frequencies were merely noted in terms of a six-grade scale during sample documentation. However, because there appeared to be a temporal pattern in their distribution throughout the stratigraphies of different mires, systematic counts of the numbers of CHAT particles were later carried out. As a working hypothesis, we suggest that charcoal and tar frequency could reflect climate at the time of peat formation and particle deposition, i.e. a low frequency would indicate a cold and wet climate with dominant cyclone activity, and a high frequency would occur under warmer and drier conditions with more frequent convective cell weather types. In this article we describe the various types of CHAT observed, and explore fire frequency implications in terms of our working hypothesis.

METHODS

Sampling sites

The geographical locations of the eleven peat stratigraphies investigated here are shown in Figure 1 and Table 1. Most of the cores originated from open raised bogs surrounded by forest which were remote from presently and previously cultivated areas, so that the incidence of CHAT arising from agricultural activity was likely to be small. However, one of the bogs (Hjortemossen) is situated in the central part of a vast agricultural area. This bog was previously surrounded by one of the largest fen areas in southern Sweden, but only the central raised bog remains now because the fen was subjected to the most extensive drainage operations in Swedish agricultural history during the 1930–40s.

Sampling and sample treatment

Because the main project involved a search for loess particles, volcanic tephra and extraterrestrial particles such as micro-meteorites and micro-spherules (Franzén & Larsson 1998, Franzén 2002, Franzén 2006, Franzén & Cropp 2007) down to the size of sub-microns, sample treatments aimed to extract the mineral particle fraction from the peat. Our procedure took much time to develop, and is described in detail here.

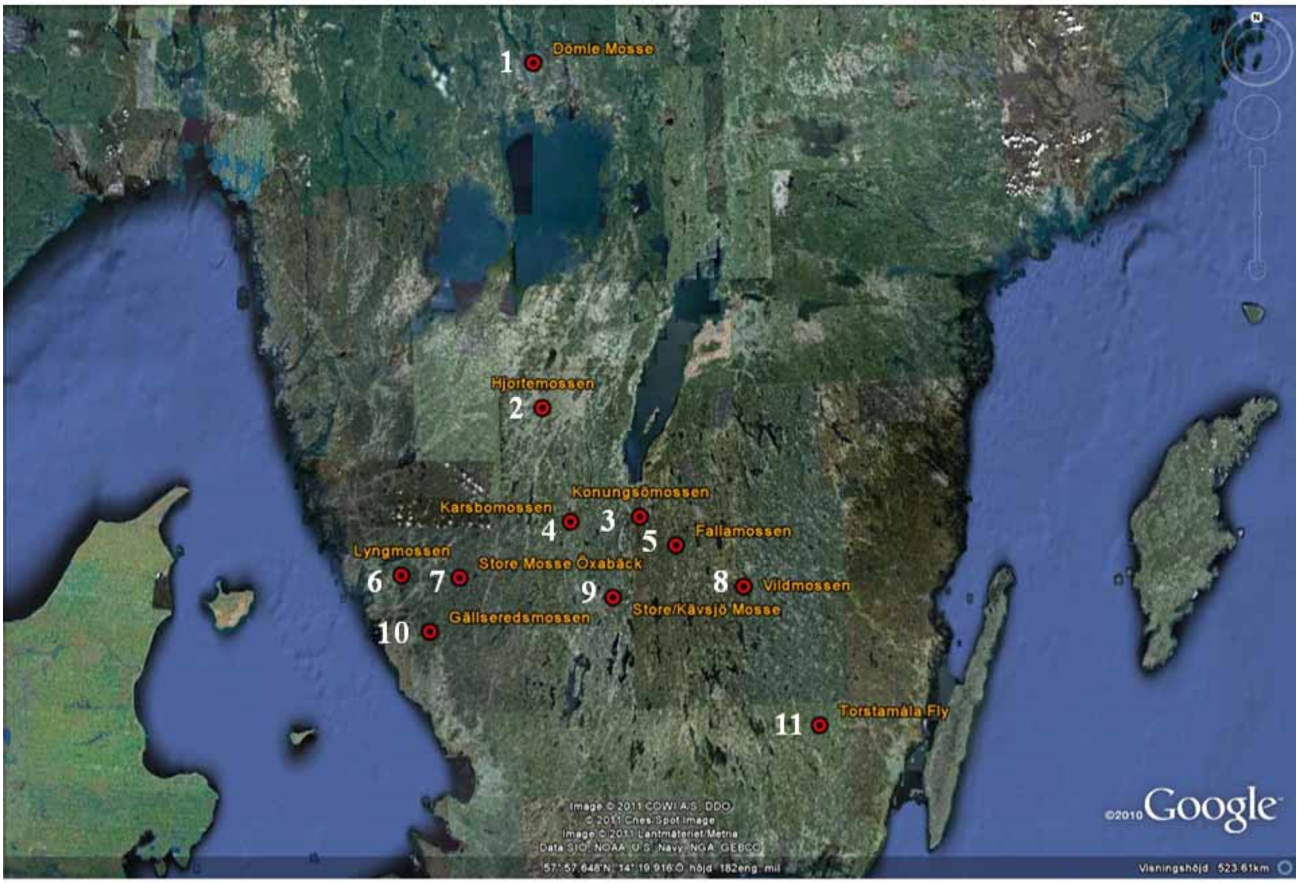


Figure 1. Southern Sweden showing the eleven sampling sites. For the exact locations of sites, see Table 1.

Table 1. Sampling sites with exact location co-ordinates and altitudes.

Sampling site		Latitude	Longitude	Altitude (feet)	Altitude (m a.s.l.)
1	Dömle Mosse	59°34’18.63”N	13°25’58.50”E	202	62
2	Hjortemossen	58°06’57.04”N	13°29’32.73”E	651	198
3	Konungsömossen	57°38’42.09”N	14°14’52.93”E	740	226
4	Karsbomossen (Komosse)	57°37’57.39”N	13°42’36.49”E	1052	321
5	Fallamossen	57°32’11.61”N	14°31’17.31”E	1157	353
6	Lyngmossen	57°24’39.78”N	12°22’51.07”E	230	70
7	Store Mosse Öxabäck	57°24’17.35”N	12°49’51.55”E	399	122
8	Vildmossen	57°20’46.46”N	15°02’48.46”E	759	231
9	Store Kävsjö Mosse	57°17’56.52”N	14°01’28.27”E	552	168
10	Gällseredsmossen	57°10’35.20”N	12°35’51.10”E	335	102
11	Torstamåla Fly	56°44’48.27”N	15°36’05.53”E	549	167

Because a bog normally starts to develop in a moist depression which forms the central point from which the mire expands laterally (Franzén 1994, Franzén *et al*. 1996), sampling was always performed on the central summits of the peatlands in order to obtain the longest records possible. Samples were taken with a stainless steel Byelorussian peat corer (50 × 500 mm), or (later) with a carbon fibre composite (CFC) corer (60 × 500 mm) (Franzén & Ljung 2009), and cut into 5 cm pieces, allowing a temporal resolution of *ca.* 50 years (peat accumulation rate is *ca.* 1 mm *per annum* (Franzén 1985)). In the laboratory, the samples were placed in deionised water, each with a spoonful (approximately 10 g) of sodium hydroxide (Puriss) added, and then stirred. The samples were left overnight and processed the following day.

Subsequently, the samples were washed through an ordinary 0.5 mm metal sieve with running deionised water, and collected in the sieve bottom. After approximately five minutes of sedimentation in the sieve bottom, the fluid was decanted and the residue put into a one-litre beaker which was then filled up with deionised water. Using a water suction unit attached to a plastic tube and a thin plastic nozzle, the main part of the “raw sample fluid” was removed, working from the top downwards, until approximately 2 cm remained. The beaker was then emptied through a 0.125 mm textile filter with a specially designed filter holder into a smaller (250 ml) beaker. The beaker, with the filter unit inside, was floated in an ultrasonic bath with the aid of a simple home-made polystyrene foam “island” and kept there for about ten minutes. The filter unit was then lifted up and, after about ten suction decantings and refillings with de-ionised water, the liquid was diluted sufficiently for the final decanting. Leaving around 1 cm of water in the 250 ml beaker, the water was slowly rotated by hand to gather the mineral particles in the middle. After sedimentation a final suction was made from near the beaker wall to leave only a few millimetres of water. The sediment was transferred to a 60 mm glass petri dish (polythene plastic dishes are unsuitable because of their static electricity properties). The petri dish was filled up with deionised water, and a final round of sedimentation and suction was carried out. Finally, with about 5 mm of liquid in the dish, it was rotated as described above to concentrate the heavier material in the middle. This caused the less dense organic material, such as CHAT, to spread out evenly over the bottom of the dish. Most of the remaining liquid was then sucked off from near the edge of the dish, and the sample was put in the oven (105 °C) to remove residual water.

The particles in the petri dishes were studied under a stereo light microscope (ZEISS STEMI SV8). The quantification of CHAT particles was performed using a 1 cm grid drawn on a sheet of paper placed underneath the petri dish. The numbers of charcoal and tar particles in the central square centimetre were counted. Selected specimens were picked up using the finest insect needles available and mounted on aluminium stubs with double-sided adhesive carbon tape for further study by SEM (ZEISS DSM 940). Gold plating (3 × 90 seconds) (BAL-TEC SCD 005 Sputter Coater) gave the best photographic results. Images were stored electronically using LINK Isis Auto Beam software and edited in Corel Photo-Paint.

The filtering procedures removed all particles coarser than 0.125 mm, including CHAT. The possibility that large autochthonous charcoal particles were discarded during the preparation procedure cannot be excluded. However, for this study, careful notes were taken during sampling and pre-analysis on any visual evidence for the occurrence of charred material.

Dating

One part of the main project from which the results presented here arose is a study of variation in geochemical composition, including Rare Earth elements (REE), within peat stratigraphies (Franzén 2006, Franzén & Cropp 2007). The same sequences can be used to investigate the spatial and temporal variation of atmospheric deposition throughout the whole period of peat formation. The transition from minerotrophy to ombrotrophy in bog profiles developed from fens, carrs and swamps is clearly defined by e.g. the lanthanum (La) signal.

Special attention was paid to ombrotrophic sites, i.e. raised bog deposits, because they are not affected by the influx of mineral water from the surroundings. Peat ash dissolved in Aqua Regis was measured using an Inductively Coupled Plasma Mass Spectrometer (ICP-MS). The results from this part of the study are not presented here, but supra-regional geochemical lead (Pb) horizons were used to provide temporal correlation between the different bogs in cases where datings were sparse (Franzén 2006, Franzén & Cropp 2007). The Holocene variations of Pb in Sweden are well known (e.g. Renberg *et al*. 2001), and one good stratigraphical Pb marker is the so-called Roman Peak centred at 2000 BP (Franzén 2006). Other stratigraphical markers that can be used for temporal correlations between the different peatlands are the volcanic glass horizons. Three major volcanic events can be detected in all of the bogs investigated, i.e. Hekla 4 (*ca*. 3950 cal ^{14}C BP), Minoan Santorini (*ca.* 3650 cal 14C BP) and Hekla 3 (*ca.* 3150 cal 14C BP) (see Swindles *et al.* 2010). A large number of peat samples from different stratigraphical levels have been ^{14}C-dated at the Uppsala AMS laboratory. For calibration, we used CALIB© (Stuiver & Reimer 2000).

Inter-sample interpolation within the individual peat cores was carried out by fitting an n-order polynomial regression curve. All peat samples could be assigned an interpolated mean age from the best-fit (n-grade regression) equation. The observations from all mires were then assembled into one file which was sorted in order of increasing interpolated age to produce a final graph showing 11-point and 55-point running means of CHAT particle frequency.

Spectral analysis

The spectral analysis was carried out on the data from all eleven mires combined into one large file. We employed the REDFIT procedure, introduced by Schulz & Mudelsee (2002), to test for cyclical patterns in the CHAT time series. REDFIT is specifically adapted to unevenly-spaced time series and allows testing of the significance of peaks in the spectrum against a null hypothesis of a red-noise background estimated using an AR(1) process. An AR(1) process is a linear Gaussian first-order autoregressive process that is used in time-series analysis to model and predict the behaviour of natural phenomena. The REDFIT technique avoids the significant bias in the form of reddening of the spectrum that results from interpolations in the time domain to artificially create equal sampling intervals. The procedure involves a test run to assess whether or not the spectrum is consistent with a red-noise model. As the lower level for the detection of non-AR(1) components in the time-series, we used the false-alarm level of $(1 - 1/n) * 100$ % (Thomson 1990), where n is the number of data points in each WOSA (Welch Overlapped Segment Average) segment inherent in REDFIT (1174 in this case); this is the maximum spectral amplitude expected if the time series were generated by an AR(1) process (Schulz & Mudelsee 2002). In our application, the false-alarm level (FAL) was 99.83 %, and a Welch spectrum was used. With this significance level the risk of identifying spurious peaks is eliminated. The spectral analysis was based on data extending back to 6500 cal ^{14}C BP.

RESULTS

CHAT particle types

Although larger macroscopic pieces of charred wood were sometimes evident during sampling, most of the particles found were microscopic, around 5–100 µm in diameter. Two distinctly different groups of particles, with great morphological variation, were distinguished: (A) charred fragments of plant tissue and (B) objects formed from tar. The former category includes charred coniferous pollen grains and microscopic pieces of charred wood and herb tissue. Particles from the tar group were classified into the following five sub-groups: (B1) large spherules with an inverted golf-ball-like surface; (B2) large spherules reminiscent of mulberries; (B3) small, perfectly spherical to elongated fusiforms and droplets with a glossy appearance; (B4) irregularly shaped tar fragments; and (B5) solidified tar foam. The differences were obvious from both light microscope and SEM examination (Figures 2–7).

(A) Charred pieces of herbaceous plants and other charred plant tissue

This group has very heterogeneous morphology depending on plant species and the type of tissue that was charred. Normally the skeletal cell walls stand up as elongated laths/rails (Figures 2c, d, f). The highest frequency of charred plant tissue is recorded in the basal peat layers that were formed under minerotrophic conditions, i.e. in fens or swamps. Even if microscopic charcoal particles may originate from sources outside the mires, the charred plant remains found in the peat are most likely to be of local origin, resulting from fire-prone withered reeds, sedges, grasses and other vascular plants which are common in these biotopes. At higher stratigraphical levels, i.e. the top metres of ombrotrophic peat where *Sphagnum* dominates, specimens from this group are less frequent, representing species such as *Eriophorum vaginatum, Juncus caespitotus* and *Rhyncospora alba*.

(B) Particles formed by distilled organic material i.e. tar

We assume that tar particles are most likely to have been formed by the burning of coarse plant material such as trees and bushes. Because such coarse plant material is normally rare in open raised bogs, we further assume that the closest originating areas are mire margins, but they may also have much more distant sources.

(B1) Large spherules with uneven surfaces

These objects are amongst the largest particles observed in the stratigraphies. They can be quite irregular but most of them appear to be rather perfectly ball-shaped (Figures 3a, b, c), or to take the form of elongated or irregularly shaped potatoes (Figures 3a, e). In some rare cases, dumbbells were observed (Figure 3f). The surface texture is normally rough and in some cases resembles an inverted golf-ball surface (Figures 3b, c), Large

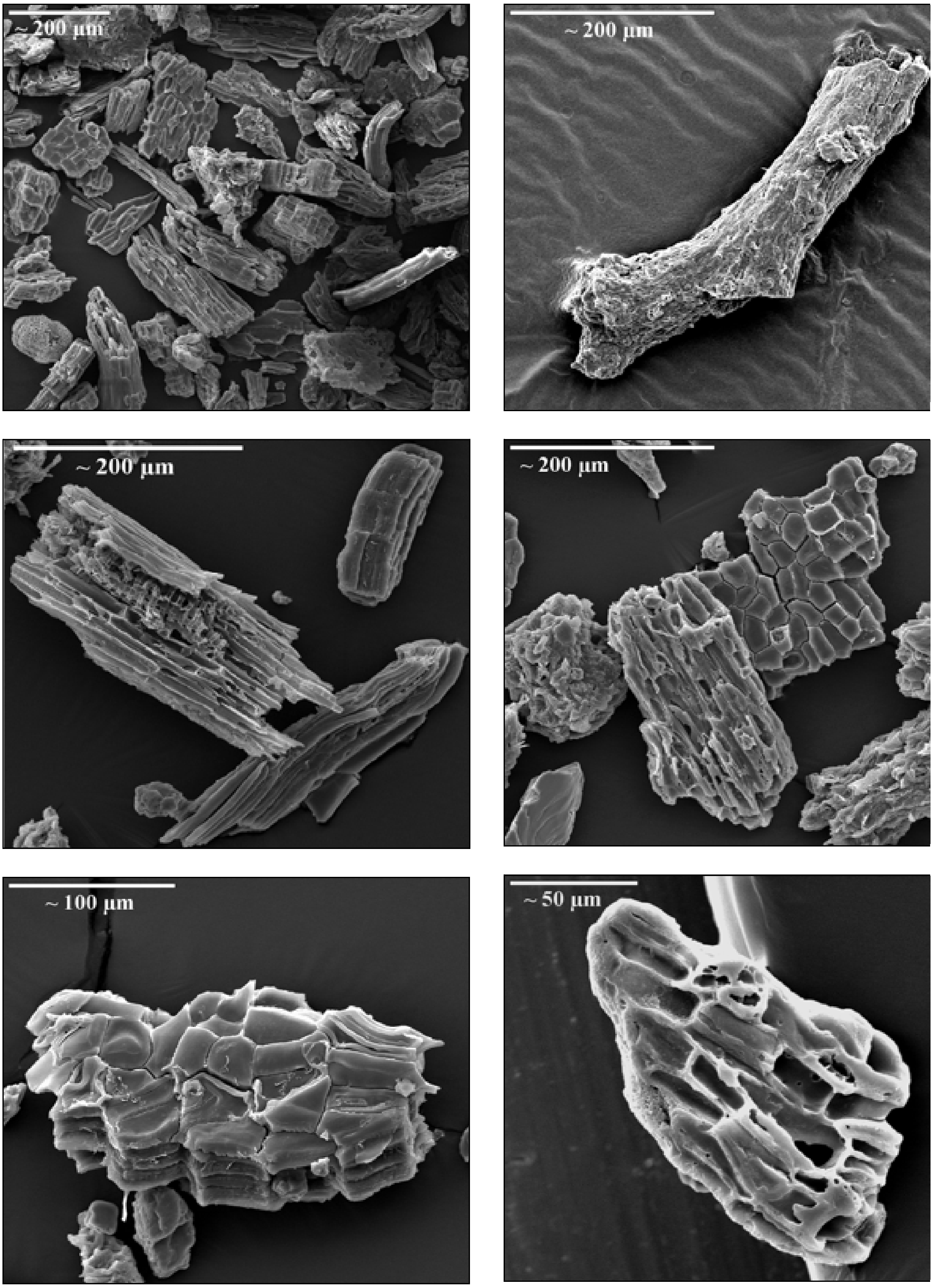


Figure 2. Fragments of charred plant tissue.

holes or cavities are sometimes found on their surfaces (Figure 3d), and transitions to Category B3 occur (Figure 3e). The large spherules occur throughout the stratigraphies but are most commonly found in the lowest, strongly decomposed ombrotrophic peat layers (5000–4000 cal ^{14}C BP). The rough and sculptured surface indicates that they were formed from relatively low-temperature tar, and their size is suggestive of an intense and fast fire lapse with strong thermal up-winds created by the intense heat.

(B2) Mulberry shaped spherules

These round objects occur rarely in the samples, and were found in only two of the stratigraphies. Their morphological appearance is most reminiscent of a

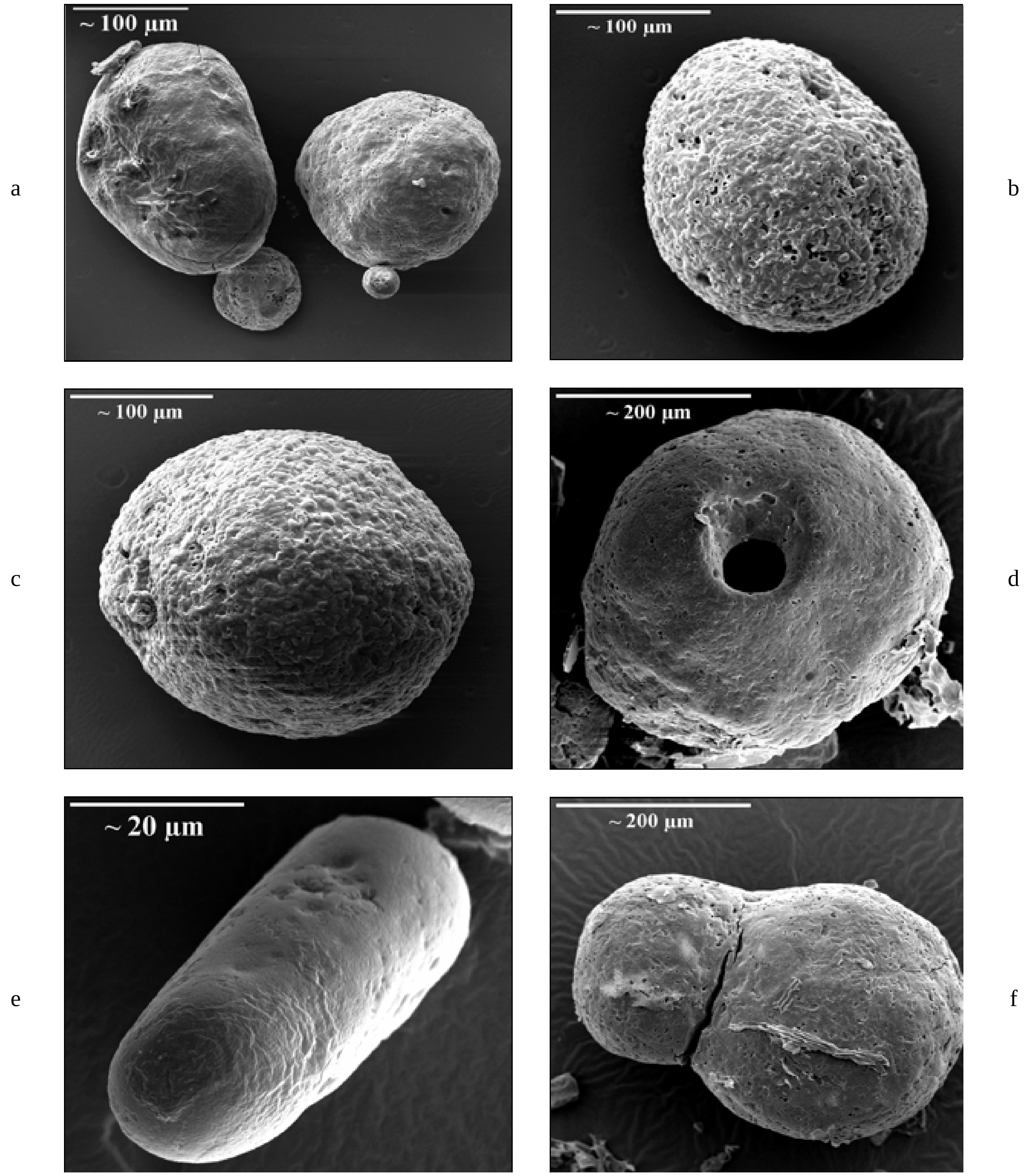


Figure 3. Large tar spherules of various forms. (c) reproduced with permission from Franzén (2005).

blackberry, raspberry or mulberry (Figures 4a–f). Small crater-like holes on the surface might have been formed by gaseous outflows (e.g. Figure 4e). These particles might have been formed as conglomerates of smaller spherules, but some other unknown process might equally well explain their genesis. Their morphology and size imply a formation history similar to that of Category B1.

<u>(B3) Tar spheres and droplets</u>

These particles may be spherical, drop-shaped, elongated ovals, dumbbells or spool-shaped

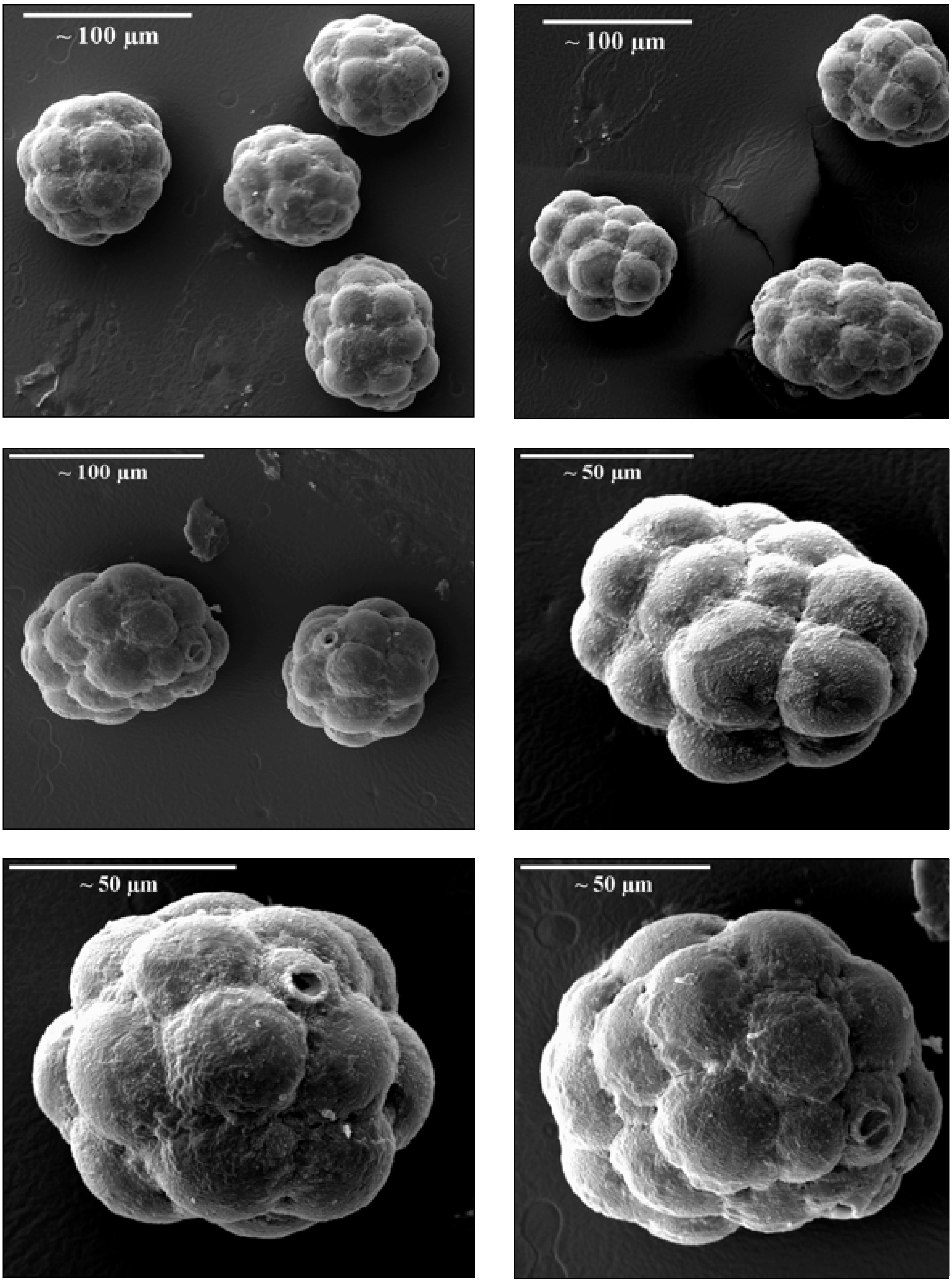
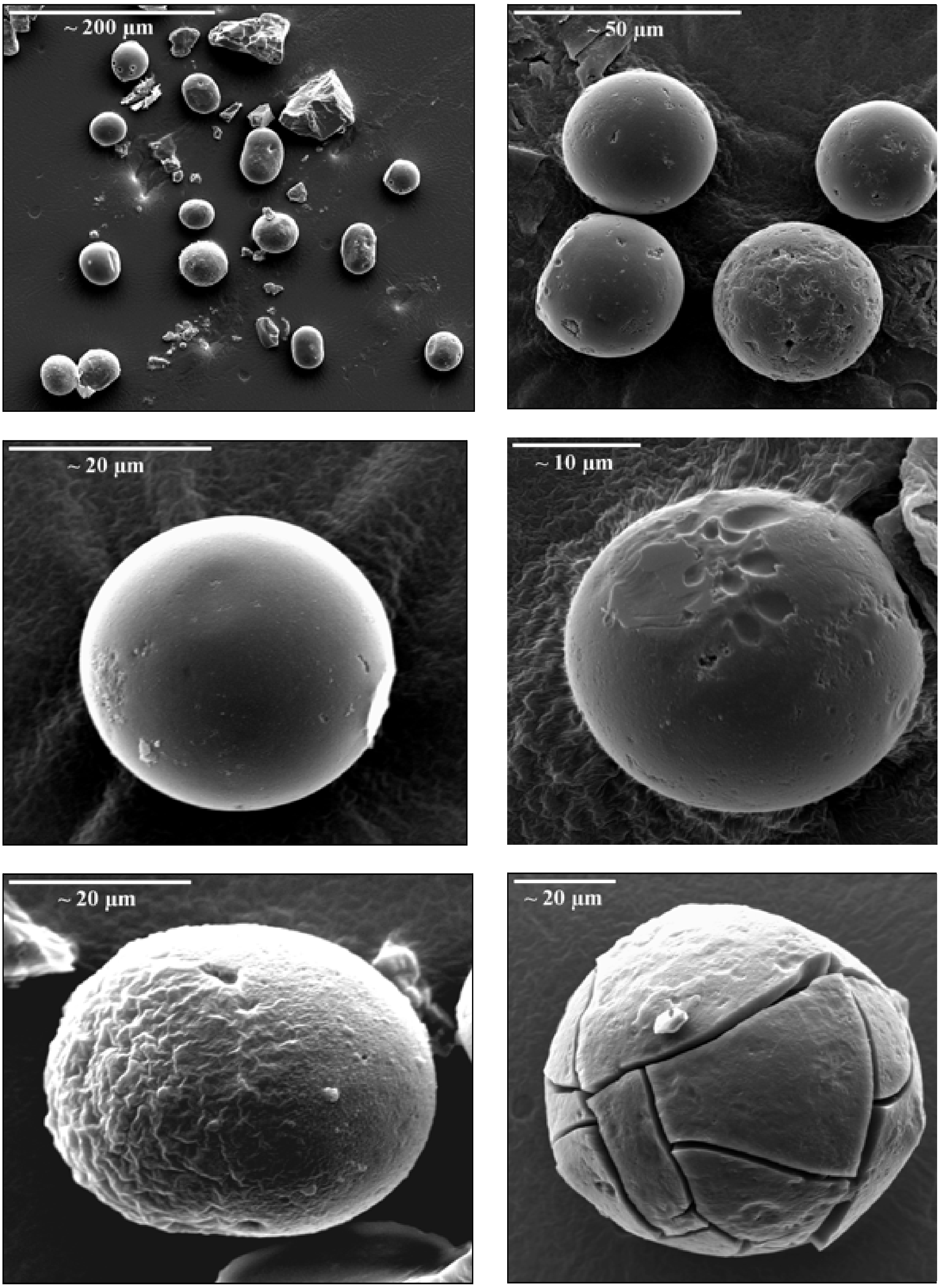


Figure 4. Mulberry shaped tar spherules. (f) reproduced with permission from Franzén (2005).

(fusiforms) (Figures 5a–r). They are easily mistaken for black iron and glass spherules (Franzén 2006, Franzén & Cropp 2007). In their simplest form they are perfectly rounded black micro- spheres with no surface texture (Figures 5b, c). They are easily recognisable under the light microscope by their glossy appearance. The variation of morphological shapes is almost unlimited, ranging from spheres to elongated fusiforms (Figures 5h–m). Both symmetrical and asymmetrical shapes occur. Sometimes they are spotted with small dimples (Figures 5g, k), which are possibly small surface



a b c d e f

Figure 5 (a–f). Black glossy tar spherules and fusiforms.

bubbles whose roofs have been eroded away. Some of these spheres are obviously hollow, i.e. solidified tar bubbles. In some cases they have been cracked by shrinkage during solidification (Figure 5f). The more far-reaching metamorphism and the much smaller size imply a higher formation temperature and both longer and higher transport. In this group we have also observed perfect drop forms, such as

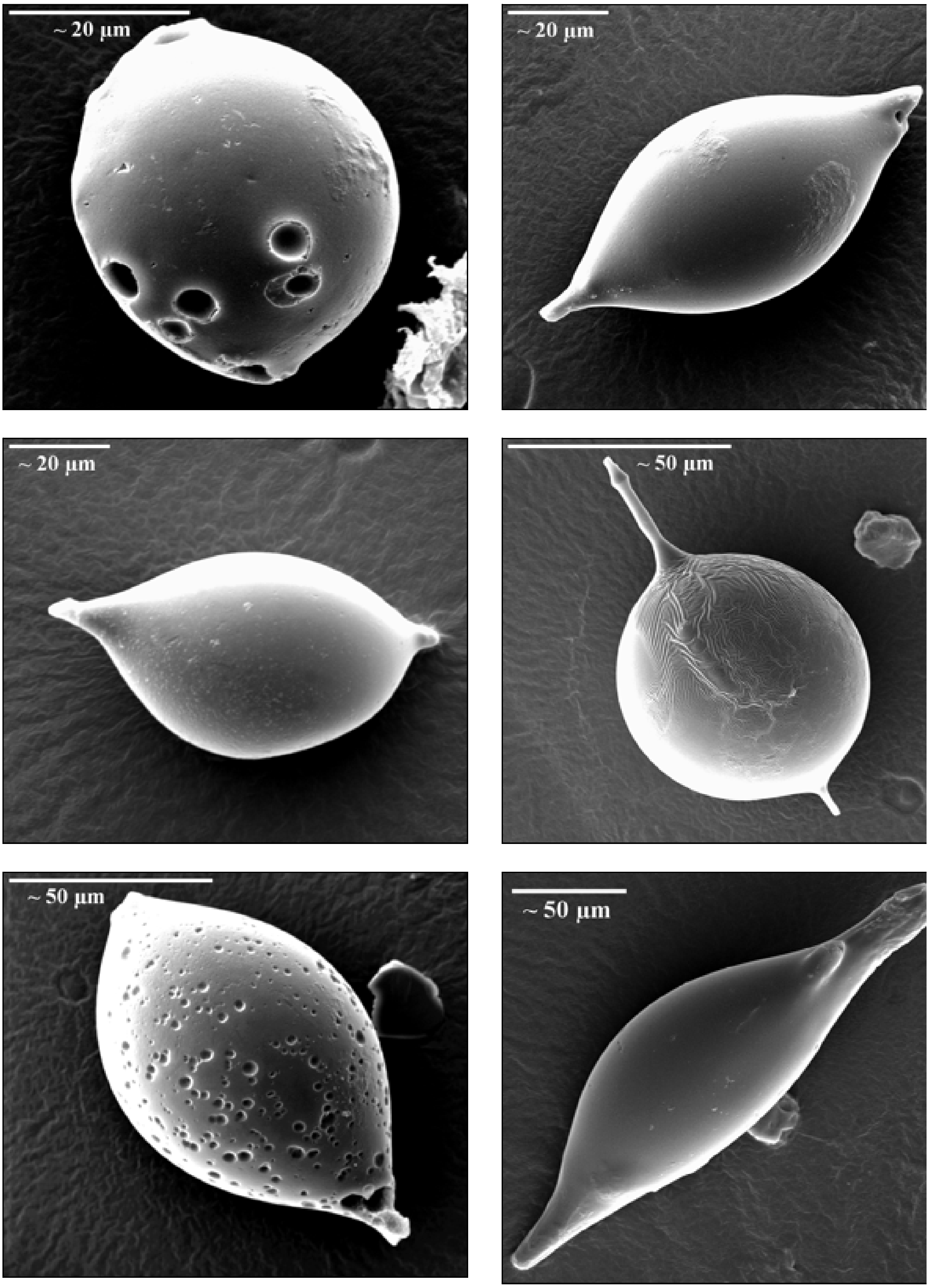


Figure 5 (g–l). Black glossy tar spherules and fusiforms. (h) reproduced with permission from Franzén (2005).

the ones shown in Figures 5j and 5o–r. The drop shape is a good indicator that the material was once a liquid, i.e. wood tar.

(B4) Irregular tar forms

Irregular tar objects (Figures 6a–e) are found only in peat layers where Type B3 occurs. Under the light microscope, they have a black glossy appearance. They are brittle and crack easily when handled. In SEM the surface structure is highly variable, with dimples and regular holes. Some of these objects, e.g. the object shown in Figure 6e, are very similar to some volcanic glass shard forms (Figure 6f).

(B5) Tar foam

Under the light microscope, these particles are black or dark brownish in colour and they look lustreless or frosted. In SEM (Figures 7a–f) they resemble

m n o p q r

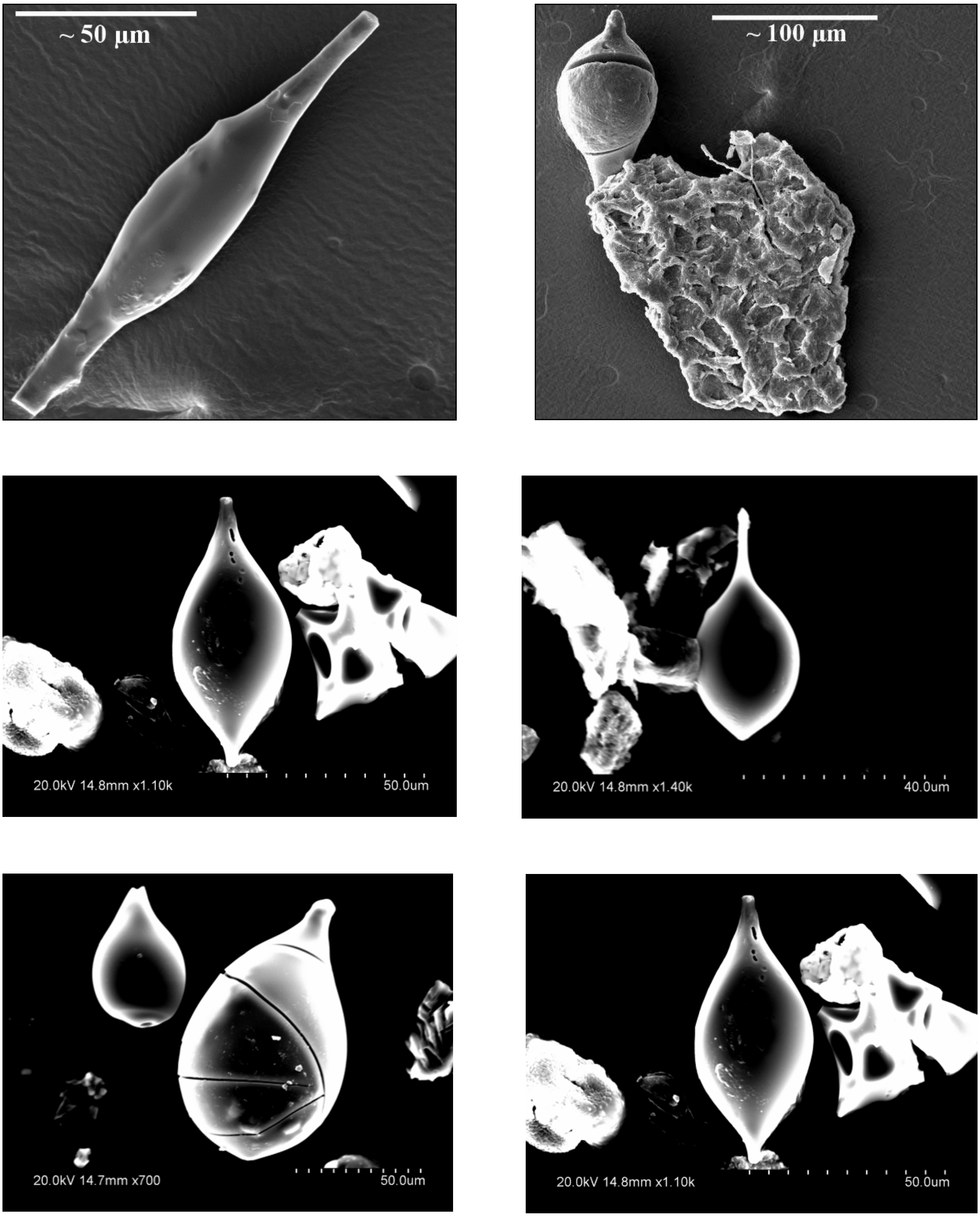


Figure 5 (m–n). Black glossy tar spherules and fusiforms.

volcanic pumice (Figures 7a–b) or plastic bubble-wrap. Closer examination shows that the whole particles are formed from microscopic bubbles of tar glued together (Figure 7d). Surprisingly, these objects have more or less the same density as the other particles. One would expect "pumice" made of tar to float and hence to be lost in the preparation procedures described above. This might actually be the case, the ones observed in the samples being heavier than normal for one reason or another. The tar foam particles occur throughout the stratigraphies and, along with the large spheres, are by far the most common of the "tar family". No matter how low the frequency of particles may be in a particular sample, one or two of these objects are always found.

a b c d e f

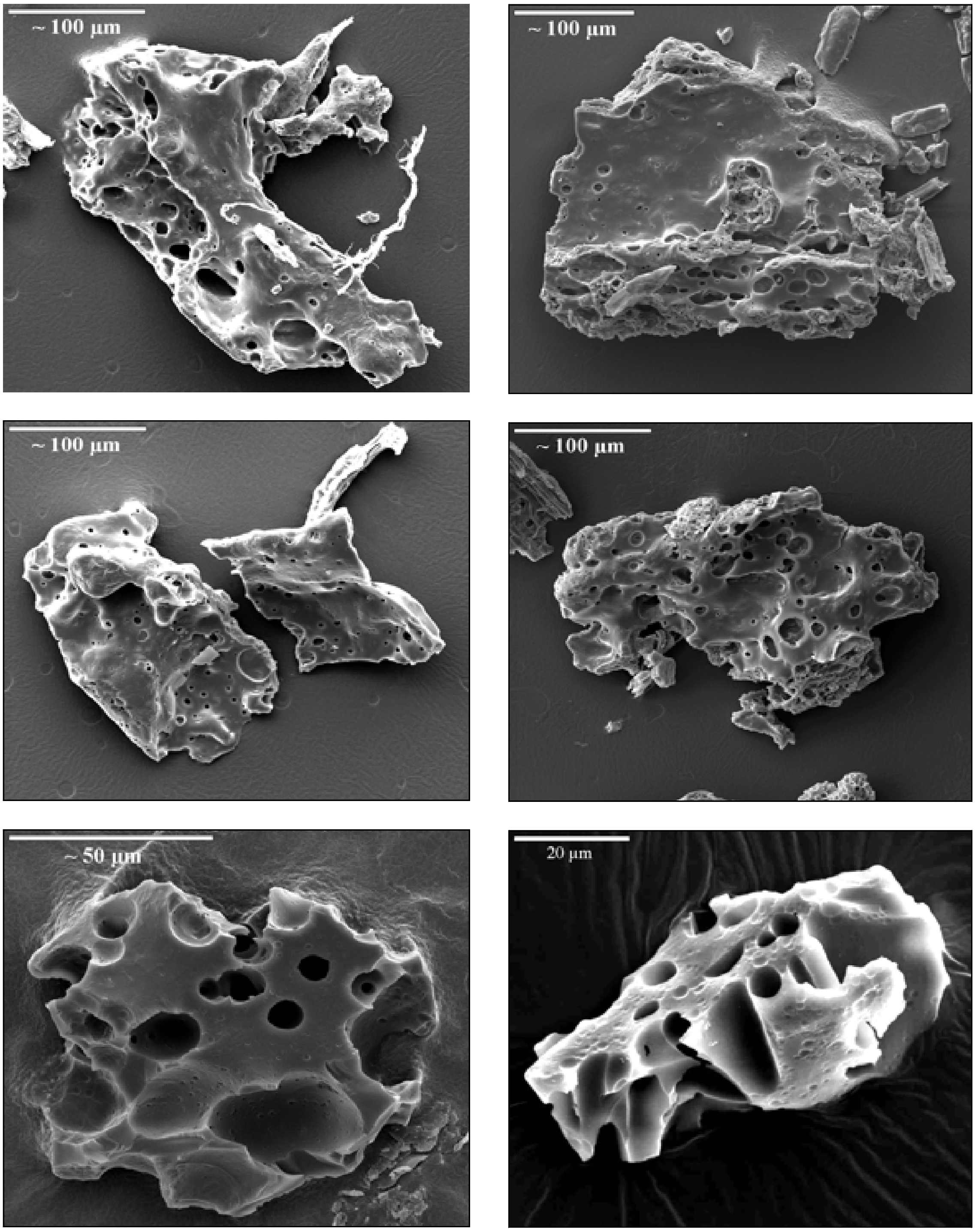


Figure 6. Irregular tar particles. Figure 6f is a volcanic glass shard (Lyngmossen, south-west Sweden, *ca* 3650 ^{Cal}BP—Minoan Santorini eruption). (e) and (f) reproduced with permission from Franzén (2005).

Minerotropy/ombrotrophy transition

Figure 8 shows the distribution of lanthanum in the stratigraphy of Gällseredsmossen, which is situated some 25 km inland from the west coast of southern Sweden (Figure 1). The transition from minerotrophy to ombrotrophy (M/O transition) is clearly visible at a depth of approximately five metres. Bogs such as Karsbomossen or Store Mosse Öxabäck (Figure 1, Table 1), that were formed directly on the mineral soil as a result of paludification, normally lack this transition and have a low REE signal overall. The top layers of all investigated bogs also show a relatively high REE signal due to modern deposition of dust created by different human activities (Franzén 2006).

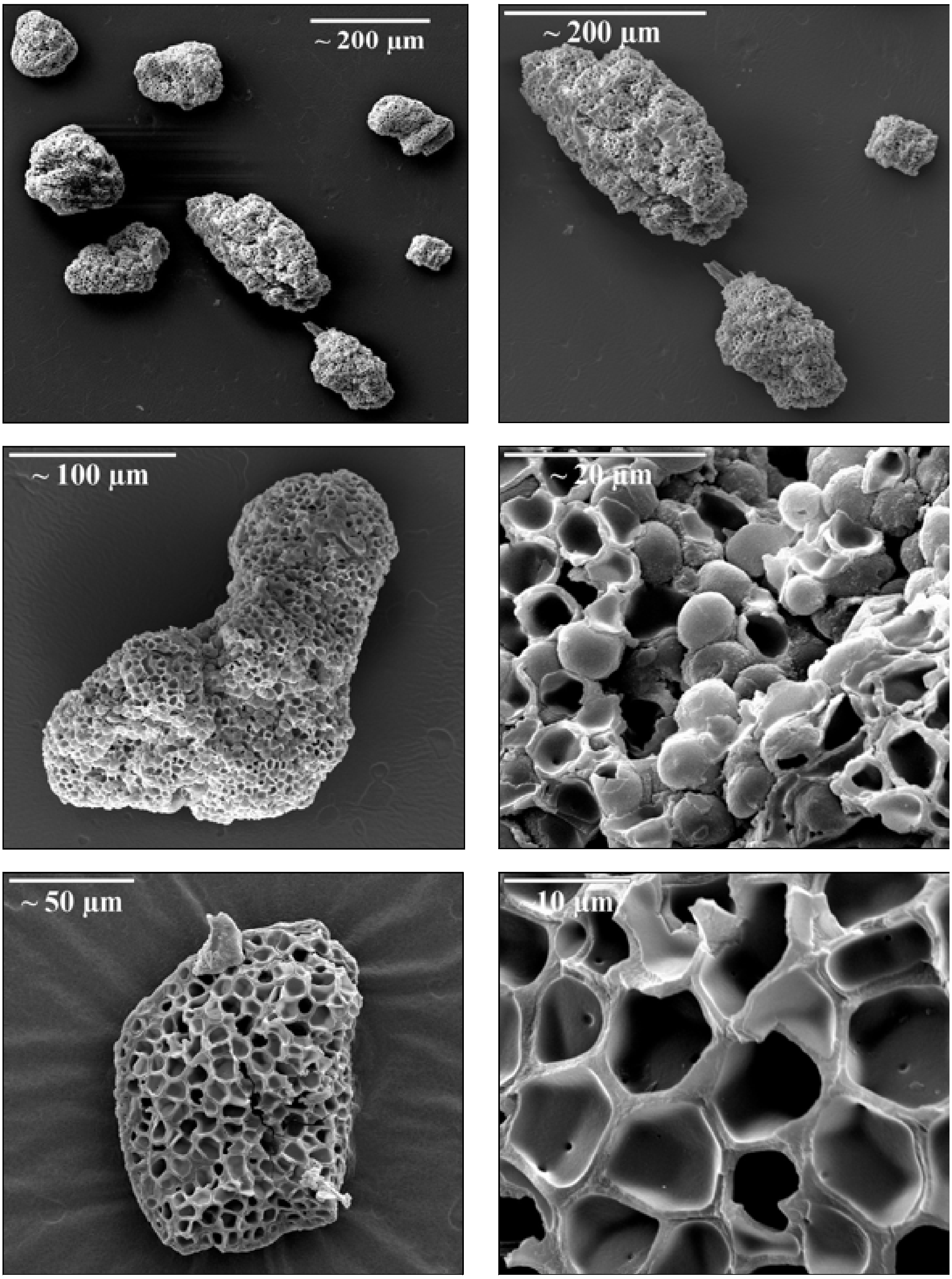


Figure 7. Objects formed by solidified tar foam. (c) reproduced with permission from Franzén (2005).

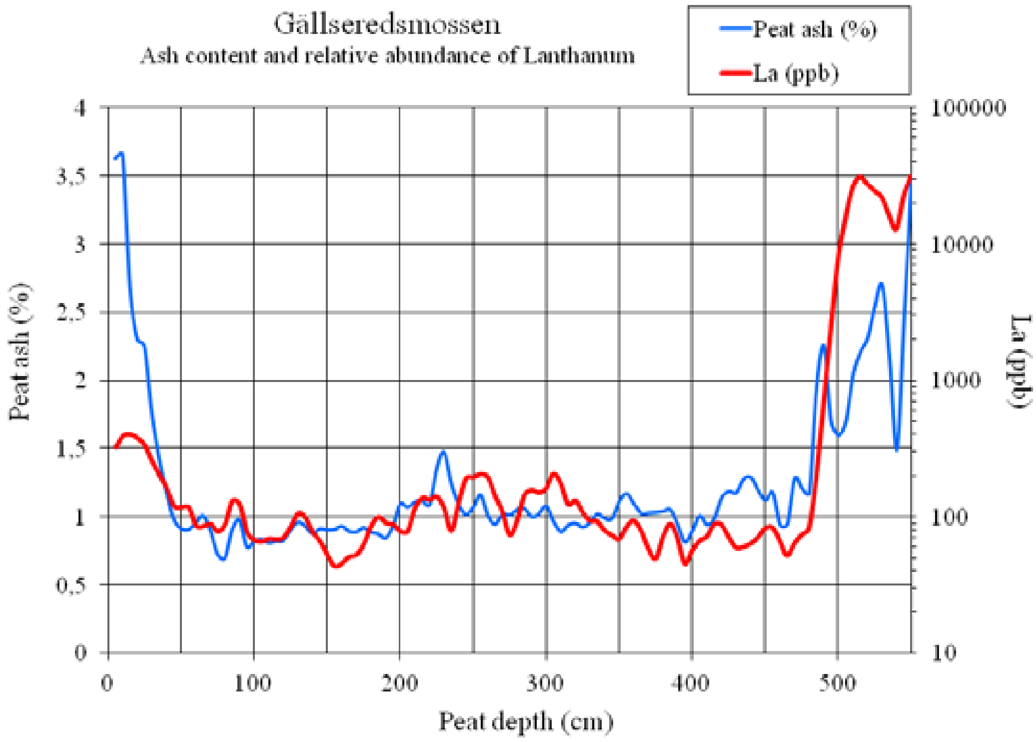


Figure 8. Stratigraphical distribution of peat ash (%) and lanthanum (La) in peat (ppb) from Gällseredsmossen, south-west Sweden (Table 1, #10). Note logarithmic scale for La.

Temporal resolution

The age-depth procedure used here is exemplified by the curve and formula for Lyngmossen shown in Figure 9, although the temporal resolution was actually derived from the single large file in which data from the eleven mires were combined. In the ideal case, and assuming that the calculated dates for the whole material were spread linearly over the whole time series, and not by using n-grade regressions, the optimal theoretical "temporal resolution" was calculated at *ca.* 4.5 years. In fact, during periods with a high rate of peat accumulation the temporal spacing (with our sampling interval) is shorter (Figure 9, less steep gradient), while during periods with a lower rate of peat accumulation it is longer (Figure 9, steeper gradient).

Fire trends

No coarse charcoal was recorded in true ombrotrophic peats, but it was more common in the lower minerotrophic peats. The largest abundance of particles, with a predominance of Type B1, was found in the lowermost peat layers, especially in the case of ombrotrophic conditions, i.e. bog peats formed directly on mineral soils.

The individual counts are presented in Figure 10. The running means of particle abundance for all eleven mires combined are shown in Figure 11. The temporal distribution of particles in the peat cores shows a rather distinctive pattern. A high influx of CHAT was generally found in the lower parts of the stratigraphies, i.e. in the lower, strongly decomposed minerotrophic fen and carr peats older than *ca*. 4000 Cal. ^{14}C BP. Charcoal was less abundant in the lower, strongly decomposed *Sphagnum* peats and in the uppermost weakly decomposed ombrotrophic peats younger than *ca*. 2500 Cal. ^{14}C BP; here, both charcoal and tar were rare and were found only in specific layers. There was no between-site difference in the lower parts of the stratigraphies, and it seems that both forest and mire fires were common throughout southern Sweden during the first few thousand years after peat formation began. The particles were mainly of local types with a high proportion of charred herbs (Type A). The sedge- and reed-dominated fens contributing to the peats formed are likely to burn during wildfires. Looking at the two main groups of particles identified here, i.e. charcoal and tar respectively, a rough division of the whole material could be made: charred plant fragments were dominant at the lower stratigraphical levels, whereas tars dominated from *ca.* 3000 Cal. ^{14}C BP. This, in turn, indicates a major climate change, from warmer to colder and wetter conditions.

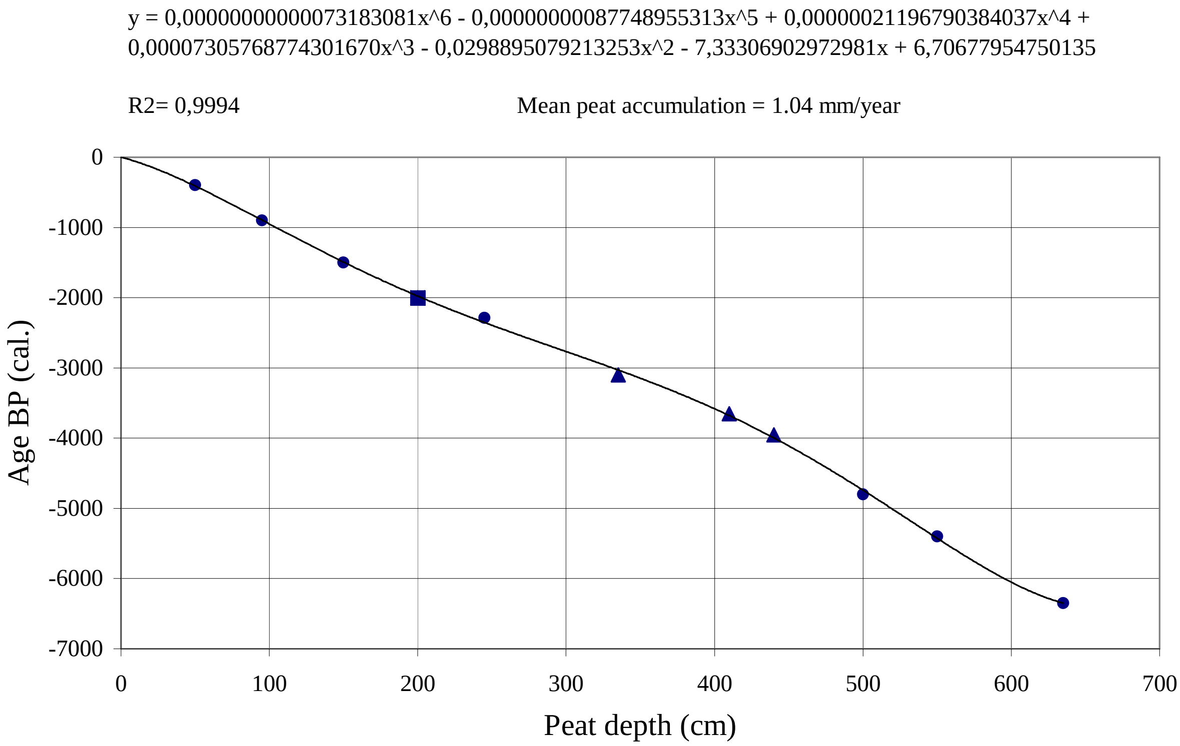


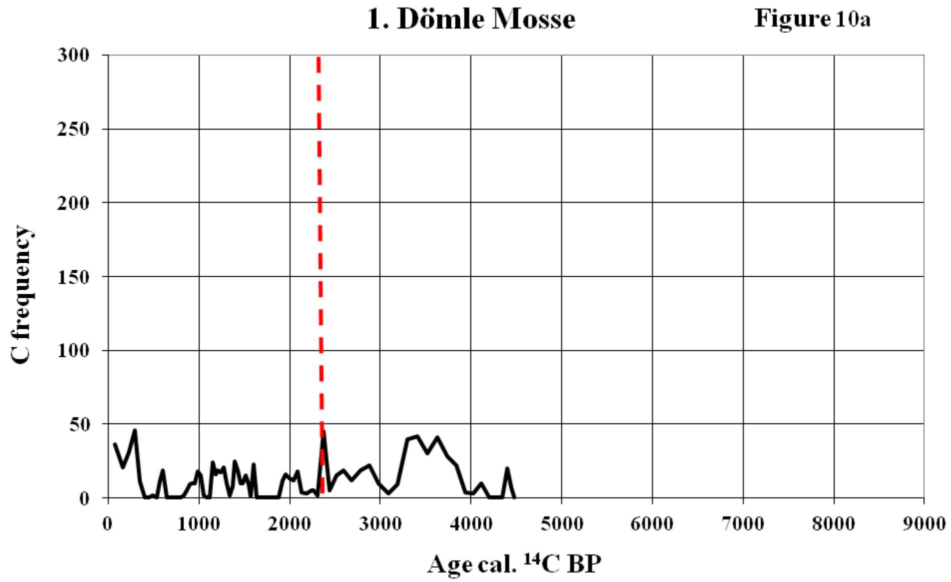
Figure 9. Datings from Lyngmossen, south-west Sweden. Dots are ^{14}C-datings, triangles are tephra horizons and the square is the Antique high peak of the Pb curve (dates are given with negative values for drafting reasons). A sixth-order polynomial fitted to these data was used to calculate age for various depths.

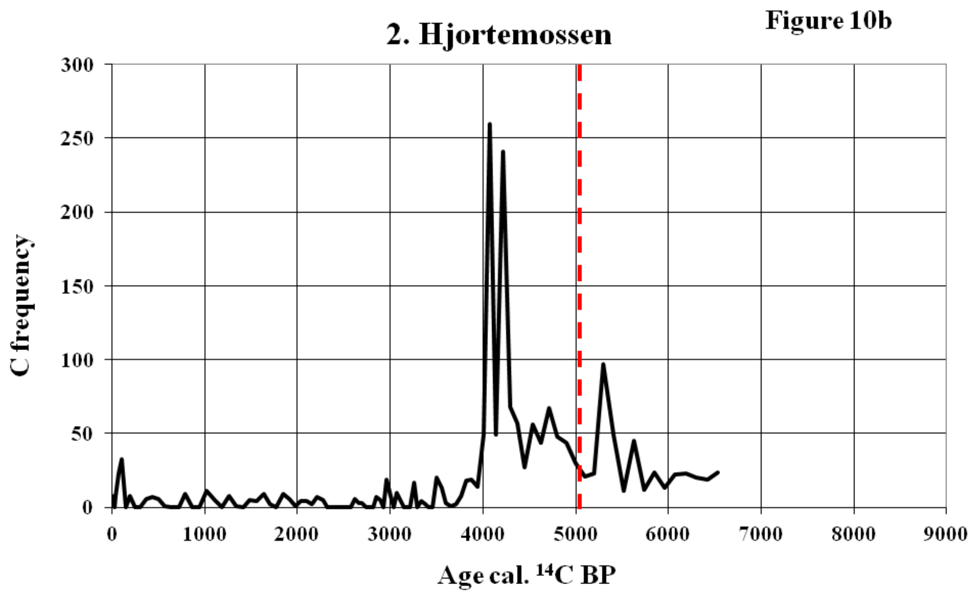
The most intensive deposition of particles, but also the largest amplitude in particle influx, was found in the lower parts of the stratigraphies. The filtered high peaks, approaching 120 particles cm^{-2} (PSC), are broken by troughs of about 30 PSC. Assuming that fires occur more frequently in periods of more continental climate with warmer and drier summer conditions, but also with a higher frequency of convective cells and thunderstorms (natural causes of fire other than lightning could probably be excluded for the south Swedish pre-anthropogenic landscape), such periods were the four distinct maxima centred around 5900, 5200, 4700, and 4000 cal ^{14}C BP. Relatively high fire peaks occurred also at 5500, 3700, 3400 and 2900 cal ^{14}C BP. From a climatological point of view, the low peaks would be indicative of a more maritime climate with frontal or orographic precipitation and fewer thunderstorms. The first particle minimum is reached around 6500 cal ^{14}C BP, a second large minimum around 5600 cal ^{14}C BP, a third around 5300 cal ^{14}C BP, and a fourth distinctive minimum around 5000 cal ^{14}C BP. A broad minimum covers the period 4200–4500 cal ^{14}C BP after a rather steep decline from the 4700 cal ^{14}C BP peak. From around 2700 cal ^{14}C BP the CHAT abundance is extremely low with values around 10 PSC. A somewhat higher abundance is recorded around 2000 cal ^{14}C BP, coinciding with the peak of the Roman warm period. The relatively high peaks in surface peats i.e. from the last *ca.* 300 years, far from built-up areas, may be attributed to local rural domestic heating or burn-beating of wetland grazing areas. The lowest values in the series are found a few hundred years after the Roman peak around 2000 BP, and a few hundred years on each side of 500 BP consistent with the Little Ice Age.

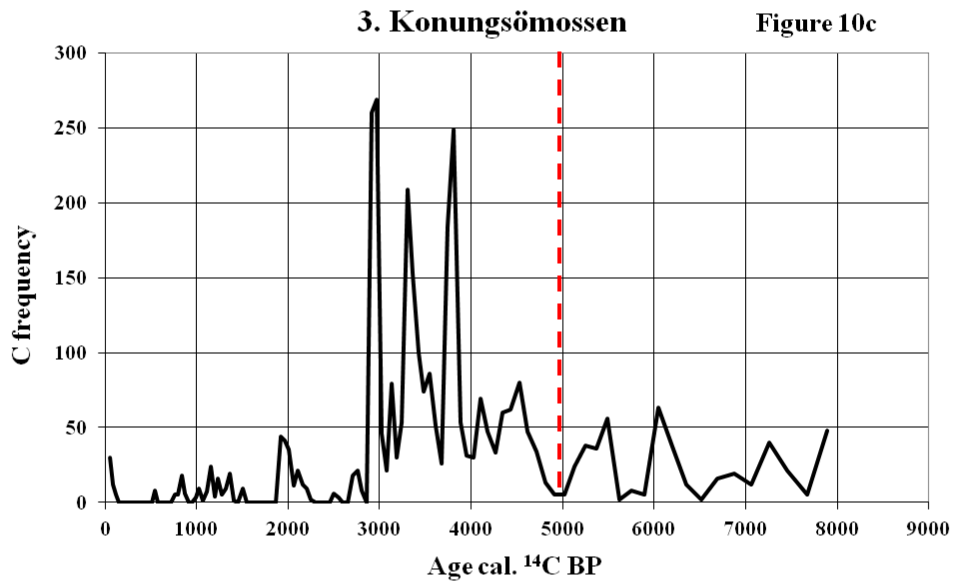
Fire frequency cycles

The first step of the REDFIT procedure involves a test run to assess whether the charcoal time series can be approximated by a red-noise model. This test gave a value of 275 with a 5 % acceptance region of 272–318, which indicates that the time series is consistent with a red-noise model (2000 Monte Carlo simulations of the red-noise spectra were used). Figure 12 shows the REDFIT spectrum on the decibel (dB) scale. Statistically significant peaks in the spectrum (above the 99.83 % FAL) are found for cycles of lengths 610, 70, 30, 21, 17 and 14 years. Thus, there is significant deviation from a red-noise stochastic process.

Figure 10 (3 pages). Temporal distributions of particles for the individual mire sites. The M/O transition (from minerotrophy to ombrotrophy), based on REE measurements, is shown as a vertical dashed red line in each case.







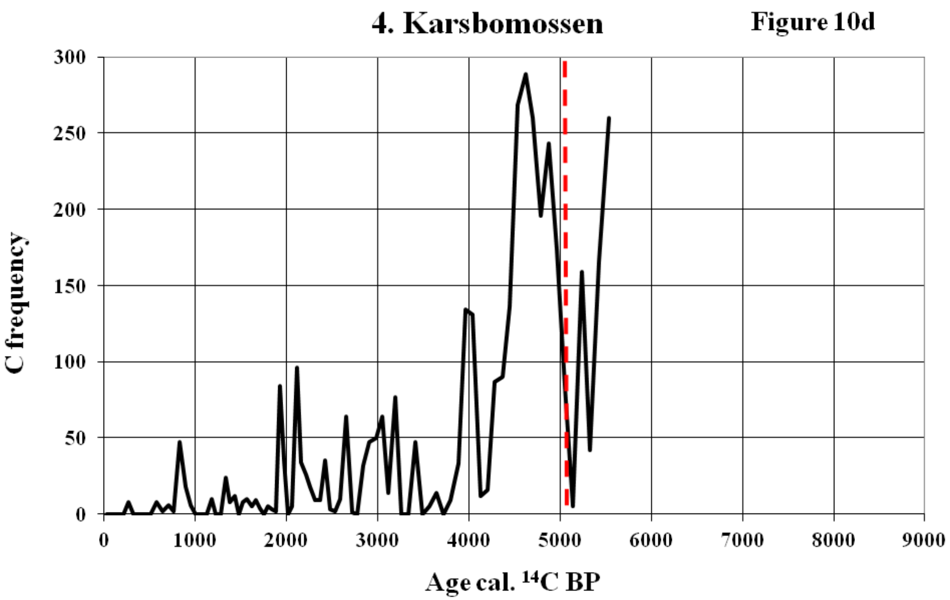


Figure 10d

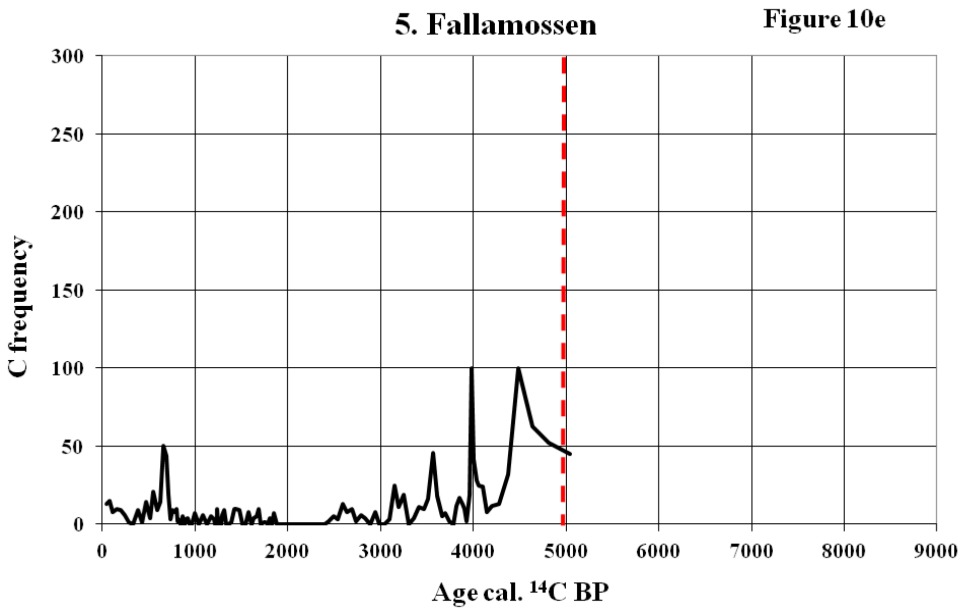


Figure 10e

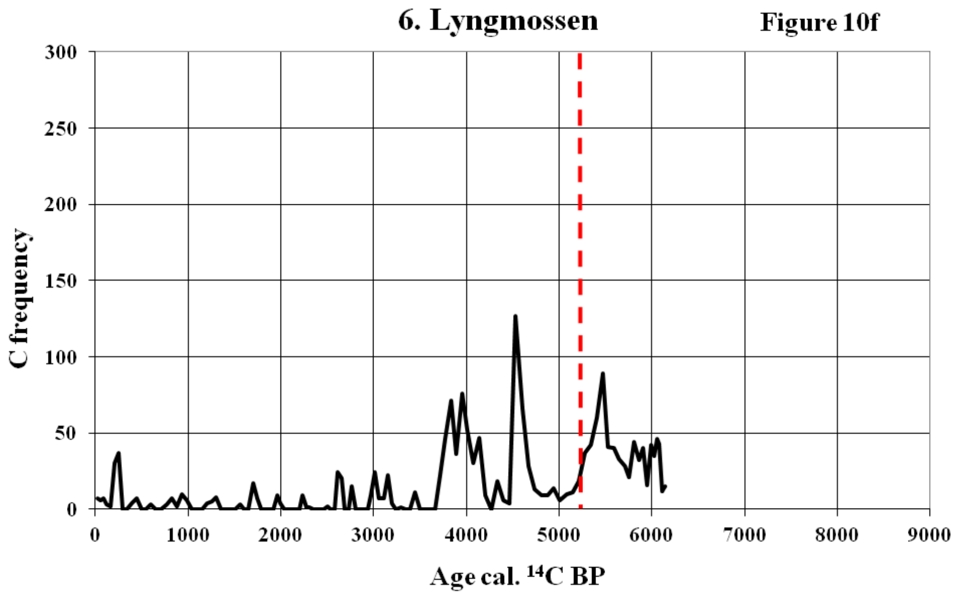


Figure 10f

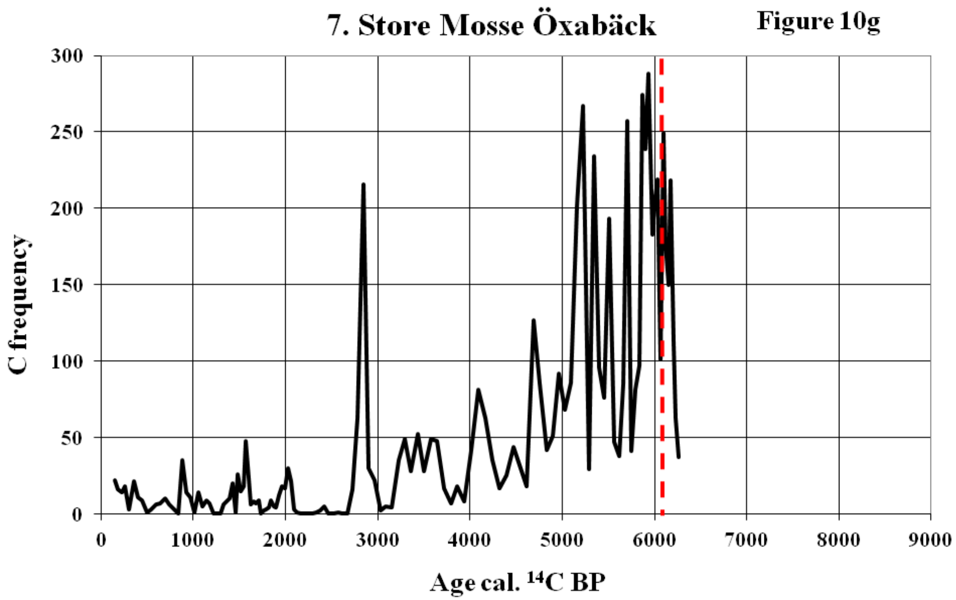
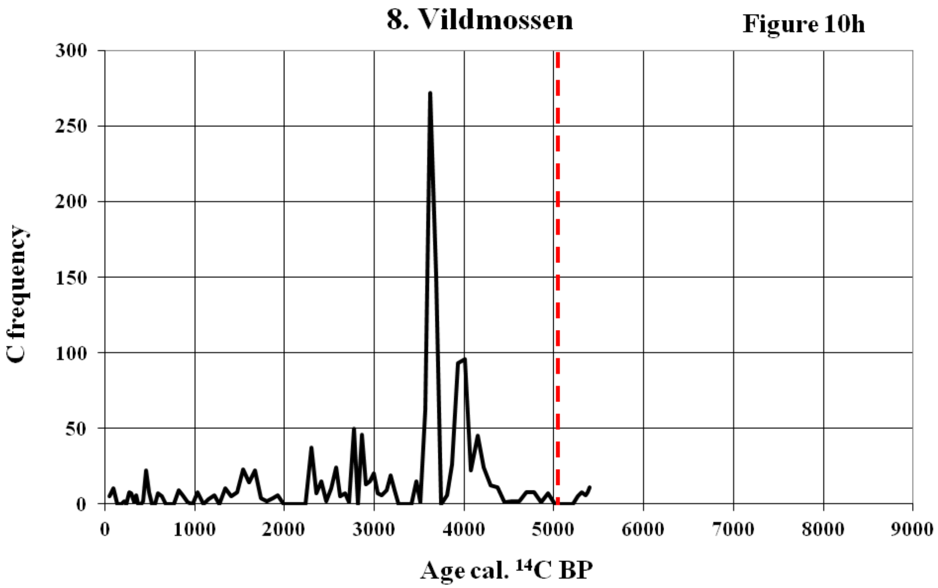
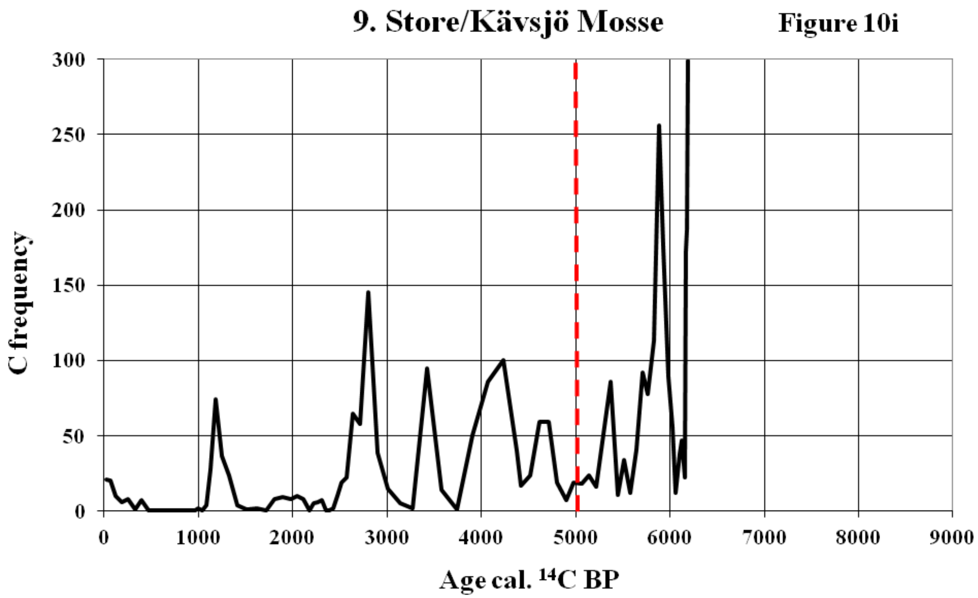
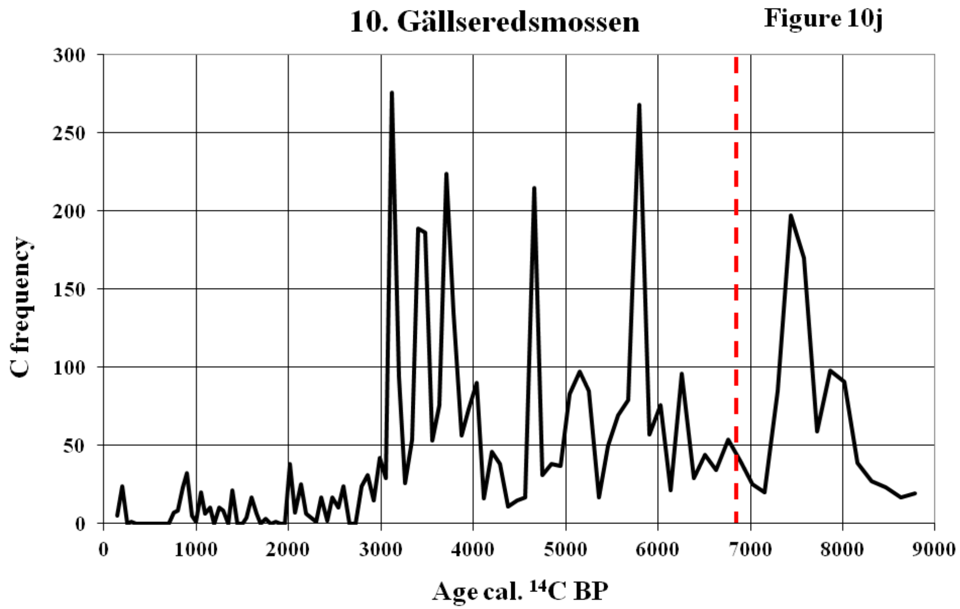
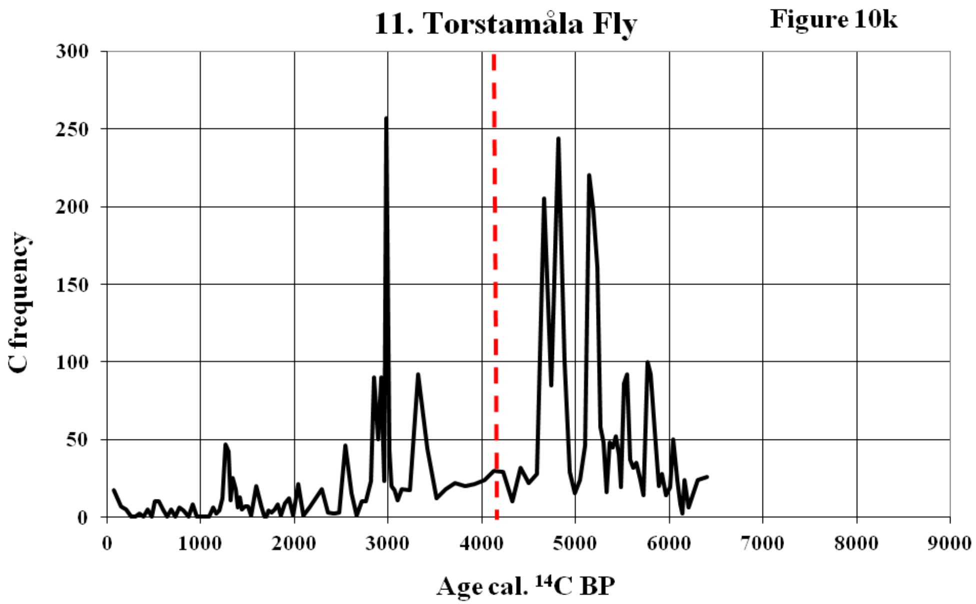


Figure 10g


8. Vildmossen
Figure 10h
C frequency
Age cal. 14C BP


9. Store/Kävsjö Mosse
Figure 10i
C frequency
Age cal. 14C BP


10. Gällseredsmossen
Figure 10j
C frequency
Age cal. 14C BP


11. Torstamåla Fly
Figure 10k
C frequency
Age cal. 14C BP

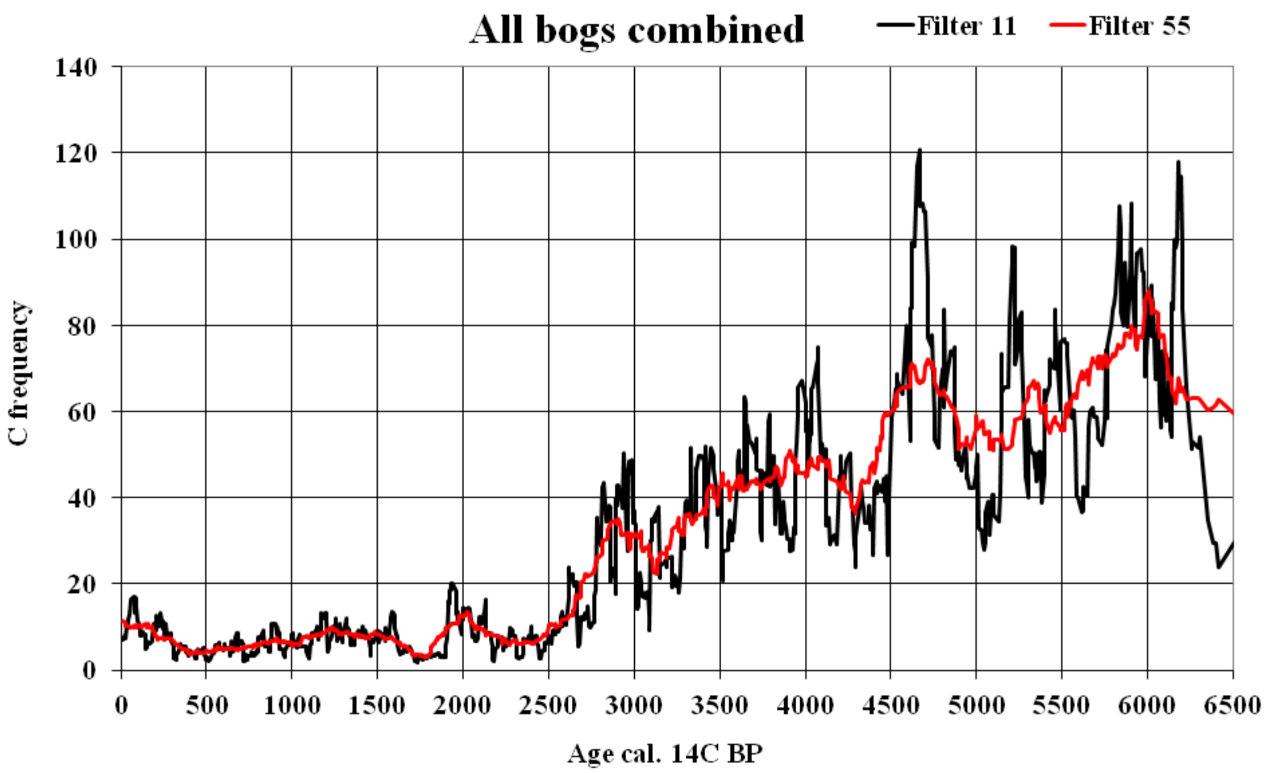


Figure 11. Temporal distribution of particles in the Swedish peat samples studied. All (n=11) mires combined. Black graph shows 11 (n) filter whereas red graph shows 55 (n×5) filter.

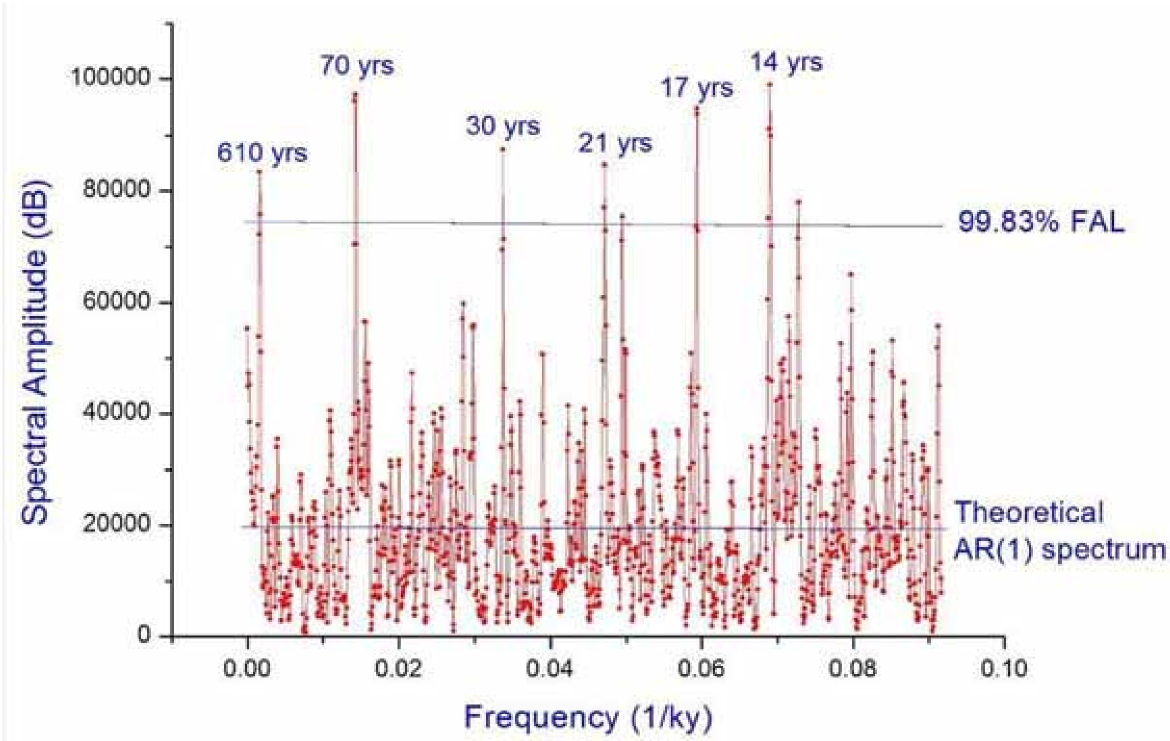


Figure 12. REDFIT spectrum (on the decibel scale) of the charcoal time series for the time interval extending back to 6500 years BP together with the theoretical AR(1) spectrum and the 99.83 % false-alarm level (FAL; Thomson 1990). The spectral peaks corresponding to cycle lengths of 610, 70, 30, 21, 17 and 14 years are above the FAL and therefore inconsistent with a red-noise process (the parameters used in REDFIT are as follows: n_{sim}= 2000, *ofac* = 4, *hifac* = 1, and *n50* = 3).

DISCUSSION

Fire trends

Our observations on the occurrence of autochthonous charcoal support the frequently debated ideas of Sernander (1910), who claimed that in most cases the initiation of bog growth was triggered by forest wildfires. Similar ideas have been presented by Pitkänen *et al.* (2001) for peatlands in Finland.

The mires investigated were all open raised bogs with scattered dwarf pines. No pine stumps were found in the upper stratigraphical levels during the coring, indicating a treeless late Holocene bog history. Whereas wildfire on bogs drained for peat cutting or other purposes is a fairly common and feared phenomenon (mainly due to the great difficulties of extinguishing such fires), wildfires are rare on bogs in their natural hydrological state due to their high water tables. After prolonged periods of drought, however, the drained and dried-out surface peat may burn for a long time at a slow rate once a fire has started. The sometimes dense layers of tree trunks and stumps in deeper peat layers (*ca*. 3000 BP), mainly in the more easterly localities of southern Sweden, bear witness to drier climates and more densely forested mire ecosystems, mainly with pine. The stumps and trunks recovered during peat cutting operations are often charred, showing that fires extended over the bogs. The onset of fires in the natural environment, with no human interference and in non-volcanic areas, is mainly due to lightning. Since lightning is associated with convective clouds, which develop mostly in summer, the periods producing large numbers of particles could be used as a proxy for climate types with these summer conditions in southern Sweden, i.e. for strengthening of the blocking anticyclone over the Baltic Sea in the manner suggested by Yu & Harrison (1995).

In a comprehensive study of Holocene humidity fluctuations in Sweden inferred from dendrochronology and peat stratigraphies, Gunnarsson *et al.* (2003) recognised the following pattern: drier conditions *ca.* 6900–6800, 4400–4200, 4100–3800, 3500–3100, 2000–1800, 1600–1400 and 650–500; and possibly wetter periods at 5600–5400, 5200–4900, 4200–4100, 3700–3500, 3100–2900, 2100–2000, 1800–1600, 1250–1100 and 450–300 BP. From lake level fluctuations in southern Sweden, Digerfeldt (1988) inferred periods of dryness (low lake levels) *ca.* 4900–4600 BP and 1800–1200 BP. Most of these periods coincide with the shifts of climate inferred from the particle data presented here. Greisman & Gaillard (2009) investigated the possible links between regional climate, fire and vegetation in southern Sweden during the early and mid Holocene using pollen, plant macrofossil and charcoal records from a small bog. High fire activity was related to dry and warm conditions around 10550, 9600, 7500–7100 and 6500 BP whereas low fire activity, due to increased precipitation, occurred *ca*. 8500–800 and 6750 BP respectively. They also found a long mid Holocene period of low fire activity, corresponding to wetter and cooler conditions, at 6350–2000 BP. Olsson & Lemdahl (2010) reported frequent fires in south-east Sweden during the middle Holocene, *ca*. 9500–1900 BP. Olsson *et al*. (2010) pointed out three major fire episodes of the early and middle Holocene from two sites in central south Sweden at 10700–10300 BP, 9250 to *ca*. 6000 BP and 1750 BP to the 19th century. These three phases are separated by periods with lower or very low fire activity. According to Olsson *et al*. (2010), fire appears to have been controlled by climate during the early and middle Holocene and by humans during the late Holocene; in addition, warmer and drier climate during the early and middle Holocene caused frequent and intensive fires.

Fire history from a Holocene perspective has also been investigated thoroughly in Finland. In a study of 98 peat cores from eastern Finland, Pitkänen *et al.* (1999) found that fires occurred most frequently during the period 7500–5000 BP, quite frequently during 5000–2500 BP, and halted from 2500 BP onwards. Despite the advance of *Picea* since about 6000 years ago, the decrease in fire occurrence was most probably attributed to a shift towards a moister and cooler climate. Tolonen (1985) considered that fires before 2000 BP were primarily natural but that more recent occurrences resulted from human activities. He identified three progressive stages of human interference, namely: temporary slash-and-burn clearance (300–700 AD), continuous intensive slash-and-burn (700–900 AD until the Middle Ages), and predominantly arable cultivation from about 1700 AD.

At a larger regional scale, proxy climate data can be compared with our record. For example, reconstructions of past changes in the hydrology of bogs in north-west Scotland indicated wet climate during the periods 5120–5070, 4020–3630, 3340–3270 and 940–800 cal ^{14}C BP, and drier periods at e.g. 4330–4120 and 1480–1340 cal ^{14}C BP (Anderson 1998). In eastern England, maximum dryness at 2000 BP was inferred from lake level fluctuations (Harrison & Digerfeldt 1993). A peat macrofossil study from northern Germany and Denmark showed cooler/wetter conditions around 2700, 1800, 1400 cal ^{14}C BP and at the onset of the

Little Ice Age around AD 1250–1350. The most marked of these periods, i.e. 2750–2600 cal ^{14}C BP, indicated a more or less catastrophical change towards wetter conditions according to Barber *et al*. (2004). From a large number of observations of terrestrial molluscs in western Europe, Rousseau *et al.* (1994) concluded that the periods around 5000 and 3000 cal ^{14}C BP were cold and wet.

Fire frequency cycles

Many forest fire cycles have been described in the literature. The validity of several of these may be questioned due to the absence of proper handling of the time-series analysis. For example, interpolations to create equal spacing of data points in an unevenly-spaced time series will inevitably "redden" the spectrum and lead to under-estimation of its high-frequency components, significantly biasing the statistical results (Schultz & Stattegger 1997). Furthermore, failure to test the significance of spectral peaks, testing their significance against a too-generous significance level, or lack of testing against a red-noise null hypothesis, may lead to the identification of cycles with little relevance. We emphasise that in the present study it was necessary to place the threshold for cycle detection at such a high level as 99.83 % (false-alarm level) in order to eliminate the risk of identifying spurious spectral peaks.

However, fire frequency is not usually calculated using spectral analysis, but with various other methods as described by Mooney & Tinner (2011). In general, the different studies of fire frequency reported in literature show that the recurrence interval has varied greatly during the Holocene. Some results are compiled in Table 2.

Table 2. Information on fire frequency, compiled from literature.

Location	**Fire frequency**	**Literature source**
West Siberian mires	2–3 times since 7–8000 BP	Turunen *et al.* (2001)
Central Siberian taiga	520 years	Mollicone *et al.* (2002)
	220–375 years	Schulze *et al.* (2000)
European taiga	50–70 years	Zachrisson (1977) Syrjänen *et al*. (1994) Lehtonen & Huttunen (1998) Sannikov & Goldammer (1996)
Muddus, north Sweden	81–90 years (most common) 110 years (mean)	Engelmark (1984)
South Sweden	20 years (last 600 years)	Niklasson & Drakenberg (2001)
Finland	80–85 years (last 3000 years)	Tolonen (1985)
	70–80 years	Pitkänen (2000)
	100–130 years (*ca.* 7500–6000 BP) 440 years (*ca.* 6000–4600 BP) 60–90 years (*ca.* 4600–1000 BP) 30–40 years (since *ca.* 1000 BP)	Pitkänen *et al.* 2001
North-east China	110–120 years	Wu-Hong (1990)
North-west Quebec (Canada)	74–112 years (last 600 years)	Bergeron (1991)
Boreal forest, Canada	50–500 years (average 100 years)	Bergeron *et al*. (2004)
British Columbia	80 years (before 1760) 100 years (after 1760)	Johnson *et al*. (1990)
Canadian Rockies	90 years (1730–1980)	Johnson & Larsen (1991)
Quebec (Canada)	500 years (last 2000 years)	Carcaillet *et al*. (2001)
Alberta (Canada)	95–185 years (last 840 years)	Larsen & MacDonald (1998)
Yellowstone (USA)	65–70 years (around 9900 BP) 330–500 years (after 9000 BP)	Millspaugh *et al*. (2000)

Most of the cycles identified in our material are not reported from other studies. However, the significant 70-year periodicity appears quite often in the literature. In addition to the sources for fire frequency listed in Table 2, Proctor *et al.* (2002) showed a 50–70 year periodicity from 1000 to 3000 BP and a slightly longer dominant frequency of 72–94 years from 1000 BP to the present, in a time series based on the growth rates of three radiometrically dated stalagmites in north-west Scotland. Schlesinger & Ramankutty (1994) identified a temperature oscillation of 65–70 years in the North Atlantic Ocean and its bounding Northern Hemisphere continents. Similar results were obtained by Kerr (2000) and Enfield *et al.* (2001), who claim that the North Atlantic sea surface temperature record for 1856–1999 contains a 65–80 year cycle with a 0.4 °C range referred to as the Atlantic Multidecal Oscillation (AMO). From a 300-year tree ring study in the interior Pacific Northwest of the USA, Heyerdahl *et al*. (2002) concluded that, at the annual scale, large fires occurred during dry years and El Niño years, whereas the incidence of small fires was independent of these factors. At longer scales the fire extent varied with precipitation and the Pacific Decadal Oscillation (PDO) was suggested as an important influence. Similar conclusions were drawn by Westerling & Swetnam (2003), who found that the El Niño-Southern Oscillation and PDO patterns appeared to modulate fire activity in the western USA during the last 300 years.

CONCLUSIONS

The temporal distribution of particles in the peat stratigraphies shows a rather distinctive pattern. High rates of particle influx/production are recorded for the earlier minerotrophic stages of mire development. We believe that these older particles are mainly of local types with high representation of charred herbs. The sedge-dominated fen types that formed these peats are likely to burn more often during wildfires than the moss-dominated raised bog communities that superseded them. The first distinct fire minimum occurred around 6500 cal ^{14}C BP, indicating more moist conditions; a second minimum is centred around 5600 cal ^{14}C BP, a third around 5300 cal ^{14}C BP, a fourth around 5000 cal ^{14}C BP and a fifth as a period of around 4200–4500 cal ^{14}C BP. All these are indicative of moister conditions, i.e. a more maritime climate with relatively mild winters and cool and moist summers with low frequencies of thunderstorms. The five distinct maxima centred around 5900–6200, 5500 5200, 4700 and 4000 cal ^{14}C BP could be explained by the converse, i.e. drier conditions during relatively short periods with a more continental climate favouring the formation of convective cells and consequent thunderstorms. The general trend towards fewer particles in the peat commencing after the 4000 cal ^{14}C BP high peak also coincides with a shift in particle type from a dominance of charred plant remains towards a dominance of particles produced by tar. This, in turn, could be interpreted as a shift from a local towards a regional fire regime, i.e. an increased importance of influx from more distant sources. The spectral analysis reveals the existence of spectral peaks corresponding to a cycle of length of 610, 70, 30, 21, 17 and 14 years, which are all clearly significant above the 99.83 % false-alarm level and which thus deviate significantly from a red-noise stochastic process. Looking at the whole time series investigated there is a general downward trend since at least 4000 cal ^{14}C BP, broken only by the most recent few centuries. This late rise can possibly be explained by the rapid increase of combustion products from different human activities (Niklasson & Granström 2000, Swindles 2010).

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