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Radiocarbon and optically stimulated luminescence dating based chronology of a polycyclic driftsand sequence at Weerterbergen (SE Netherlands)

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ABSTRACT

The chronology of polycyclic driftsand sequences in cultural landscapes has mainly been based on the combination of radiocarbon (¹⁴C) dating of intercalated organic horizons and pollen analysis. This approach, however, yields indirect age information for the sediment units. Also, as soils are dynamic systems, the pedogenetical interpretation of the ¹⁴C ages is often quite difficult.

To improve the results of radiocarbon dating, we applied fractionated ¹⁴C dating, sustained by soil micromorphology and pyrolysis-gas chromatography/mass spectrometry. The results indicate the complexity of the sources and decomposition processes of SOM, and, consequently, provide information as to why radiocarbon dates are not always reliable for the geochronology of driftsand deposits. We then performed an optically stimulated luminescence (OSL) dating study of the driftsand beds in the sequence. This approach yields direct sedimentation ages, and allows differentiating the instable (sand drifting) period from the stable (soil formation) period in each individual cycle of the sequence. Post-depositional mixing of the sands, however, may upset the reliability of the OSL chronology.

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1. Introduction

Late Weichselian aeolian coversand dominates the surface geology of an extensive part of northwest Europe (Castel et al., 1989). In the Early Holocene the area stabilized under pioneer vegetation. In the Atlantic period, a deciduous forest covered the area. In prehistorical and early historical time, forest grazing, wood cutting and shifting cultivation gradually transformed this forest into heath land. Subsequently, the use of the heath for the production of organic manure during the period of plaggen agriculture (from the early Middle Ages to the introduction of chemical fertilizers around 1900 AD) resulted in the local remobilization of the coversands, and led to major phases of sand drifting. Locally, the coversand landscape transformed into a driftsand landscape with characteristic new landforms and soils (Van Mourik, 1988). Interesting soil archives in these cultural landscapes are polycyclic driftsand sequences, geo-ecological records of a succession of cycles of alternating instable and stable phases in landscape development.

Interpretation of paleoecological information, derived from these records, requires knowledge of the chronology of the deposits. Traditionally radiocarbon dating of soil organic matter (SOM) extracted from buried humic horizons was used to date the individual cycles of the polycyclic sequences. This approach, however, has two disadvantages.

Firstly, extracted SOM from buried humic horizons has a complicated composition in terms of chemical characteristics and ages (Goh and Molloy, 1978; Ellis and Matthews, 1984; Stevenson, 1985). This must be considered when interpreting the ¹⁴C age results.

Secondly, every cycle reflects a period of landscape instability (sand drifting) and landscape stability (soil development). The ¹⁴C ages of buried soil horizons allow (at least in principle) differentiating between aeolian deposition phases, but they do not allow establishing whether periods were dominated by active driftsand deposition or soil formation.

In SE Netherlands, a polycyclic Holocene soil-driftsand sequence is well developed near the locality of Weerterbergen (Fig. 1). The profile (known as the profile “Defensiedijk”) has been investigated frequently over the past 20 years (Van Mourik, 1988; Dijkmans et al., 1992; Van Mourik et al., 1995). In this paper, we briefly summarize the previous finds for the sequence, and report on a series of new investigations (fractionated ¹⁴C dating, soil micromorphology and pyrolysis-gas chromatography/mass spectrometry) that aim at improving our understanding of the composition of SOM in buried humic soil horizons. The results confirm the complexity of SOM and illustrate why radiocarbon dating of this type of material may not always be reliable. In addition, we applied optically stimulated luminescence (OSL) dating to establish a chronological framework for the paleoecological information preserved in polycyclic driftsand sequences. The results illustrate the possibilities and limitations of OSL dating for constraining the time of sand-drifting events in the West European lowlands.

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Fig. 1. a. Location of the Weeterbergen in The Netherlands. b. Fragment of the historical topographical map (scale 1:25,000, 1900 AD) with the location of profile Defensiedijk. c. Profile Defensiedijk 1984. d. Profile Defensiedijk 2002.

2. Materials and methods

2.1. Selected profile and soil sampling.

Landforms and soils around the city of Weert are representative for the cultural landscapes that developed on chemically poor Late Weichselian aeolian coversands in NW Europe (Van Mourik, 1988). Historically, there was a close relation between the development of fmic antrosols and driftsand deposits in these cultural landscapes (Bokhorst et al., 2005). Profile Defensiedijk is situated in the Weeterbergen, SE of the city of Weert. Fig. 1b shows a fragment of a historical map (1900 AD) with the characteristic land use of cultural landscapes on chemically poor sandy soils. Around the city of Weert, arable fields (fmic antrosols) are visible. More to the west we can see the extensive heath (podzols) with complexes of land dunes

(arenosols) and the first generation of pine plantations. Profile Defensiedijk was considered as an important paleoecological record of stable and instable periods in the development of the cultural landscape. In 1984, the profile (Fig. 1c) was sampled for pollen analysis. Also samples were taken for radiocarbon dating, applied on bulk samples. In 1986 the same profile was resampled for fractionated radiocarbon dating, soil micromorphology and pyrolysis/mass spectrometry. Finally, in 2002 samples were taken for OSL dating. Unfortunately, the former profile location was seriously damaged and the new profile pit, just 4 m north of the former site, showed a similar sequence of soils and deposits, but differences in the thickness of the driftsand beds (Fig. 1d). Therefore, also control samples were taken for soil micromorphology and radiocarbon dating.

Our multidisciplinary approach involved (i) pollen analysis, (ii) radiocarbon dating of bulk material and fractionated radiocarbon dating,

(iii) micromorphological analysis, (iv) pyrolysis-gas chromatography/mass spectrometry and (v) OSL dating.

2.2. Pollen analysis (profile 1984)

Pollen extractions of samples from all horizons (vertical samples were performed using the KOH–HF–Acetolysis extraction method and pollen analysis) using the pollen determination key of Moore et al. (1991). An exotic marker was added to the samples to estimate pollen concentrations. That allowed the distinction between sin- and post-sedimentary pollen, important for the interpretation of diagrams of polycyclic sequences. Low concentrations of sin-sedimentary pollen grains occur in driftsand deposits. During stable periods, the soil surface is subjected to pollen precipitation. Due to soil fauna activity, pollen can infiltrate into the soil. The vertical distribution of post-sedimentary infiltrated pollen shows a sharp decline of pollen concentrations with depth (Van Mourik, 2001). The main research questions to answer with palynological observations were relative dating of the various driftsand deposits and whether climatic or cultural factors were responsible for the alteration of stable and instable periods.

2.3. ^{14}C dating

Conventional radiocarbon ages of bulk samples of buried A horizons were performed in order to interpret the chronology of pollen zones and sand deposits by the CIO (Centrum voor Isopen Onderzoek), Rijks Universiteit Groningen, The Netherlands, according to the methods described by Mook and Streurman (1983). The ^{14}C dates of bulk samples (profile 1984, Table 1) did not provide a clear geochronology and to improve this, the profile was resampled in 1986 for fractionated ^{14}C dating (Van Mourik et al., 1995). Based upon extractability behaviour, three specific organic fractions can be defined: the fulvic acids (FUL; soluble in acid and in lye), the humic acids (HAC; insoluble in acid and soluble in lye) and the humin fraction (HUM; insoluble in acid and in lye). The biological decomposition rate of fulvic acids is relatively high; they migrate easily through the soil profile or leach away completely. Therefore, they are unreliable for dating purposes. The biological decomposition rate of humic acids is medium high. Compared with FUL, they are immobile in the soil profiles and reliable for dating purposes. The ^{14}C age of HAC will be close to the moment of fossilization (burying) of the soil. HUM will accumulate during an active period of soil development; therefore, ^{14}C ages of this fraction will overestimate the time of fossilization of the soil. It is assumed that the differences between the ages of HUM and HAC increase during an active period of soil formation.

2.4. Micromorphology (profiles 1986 and 2002)

For micromorphological analysis, undisturbed samples were taken in Kubiena boxes for the production of thin sections (7 cm/4 cm/20 μm) (Brewer, 1976). The main research questions to answer with micromorphological observations were whether sin-sedimentary and

post-sedimentary SOM particles can be distinguished, and disturbance of fossilized soil horizons can be observed. Both objectives are related to assessing the reliability of ^{14}C dates.

2.5. Pyrolysis-gas chromatography/mass spectrometry and thermally assisted hydrolysis and methylation (THM) (profile 1986)

Analytical pyrolysis techniques split organic macromolecules into smaller fragments which can be subsequently analysed by gas chromatography coupled to mass spectrometry. The compounds identified provide a fingerprint of, in our case, the SOM composition, which in turn reveals information about the origin, fate and degradation of the organic carbon. Pyrolysis-gas chromatography/mass spectrometry was applied to freeze-dried SOM extracts of humic horizons. Pyrolysis was carried out on a Horizon Instruments Curie-Point pyrolyser. Samples were heated for 5 s at 600 °C. The pyrolysis unit was connected to a ThermoQuest Trace GC 2000 gas chromatograph and the products were separated by a fused silica column (J & W, 30 m, 0.32 mm i.d.) coated with DB-5 (film thickness 0.25 μm). Helium was used as carrier gas. The oven was initially kept at 40 °C for 1 min, next it was heated at a rate of 7 °C/min to 320 °C and maintained at that temperature for 15 min. The column was coupled to a Finnigan Trace MS mass spectrometer (mass range m/z 45–600, ionization energy 70 eV, and cycle time 1 s). Thermally assisted hydrolysis and methylation (THM) was performed by adding a droplet of a 25% solution of tetramethylammonium hydroxide (TMAH) in water to the sample, after which the sample was dried by a 100 W halogen lamp, and subsequently pyrolyzed, using the same GC and MS conditions as with (conventional) pyrolysis. With THM, hydrolyzable bonds are cleaved and the resulting carboxylic acid and hydroxyl groups are *in situ* transformed into their corresponding methyl esters and methyl ethers. Identification of the compounds was carried out by their mass spectra using a NIST library or by interpretation of the spectra, by their GC retention times and/or by comparison with literature data.

2.6. Optically stimulated luminescence (OSL) dating

Luminescence dating uses the constituent mineral grains of the sediment itself, and it allows determining the time of sediment deposition and accumulation directly (see e.g. Aitken, 1998). The profile Defensiedijk had previously been sampled for thermoluminescence (TL) dating using feldspar, but the age results lacked precision (Dijkmans et al., 1992). Optically stimulated luminescence (OSL) dating of quartz is better suited to date sediments, and it has been successfully applied to Holocene and Late Pleistocene sediments (Murray and Olley, 2002; Ballarini et al., 2003; Vandenberghe et al., 2004, 2009; Derese et al., *in press*) and soils in cultural landscapes (Bokhorst et al., 2005).

In 2002, the profile Defensiedijk was resampled for OSL dating (Fig. 1d). The profile pit was very close to the section that was sampled in 1986 for TL-analysis; the two profiles were very similar,

Table 1

^{14}C ages BP of bulk samples (sampled in 1984) and organic fractions of humic horizons (sampled in 1986 and 2002), profile 'Defensiedijk'.

Depth (cm)	Horizon	Bulk profile 1984	HUM profile 1986	HAC profile 1986	Depth (cm)	HAC profile 2002
025–027	2A	1130 ± 60	3230 ± 110	0410 ± 45	069–071	0410 ± 35
127–129	3Atop	1075 ± 30	1350 ± 50	1365 ± 25	128–130	1230 ± 35
129–131	3Abottom		1900 ± 110	1675 ± 30		
173–175	4Atop	3920 ± 40	4110 ± 90	3615 ± 35	154–156	2645 ± 40
175–176	4Abottom		4430 ± 165	3965 ± 40		
185–186	4Btop		3535 ± 80	3985 ± 35	170–172	2080 ± 35
187–188	4Bmiddle			3730 ± 35		
189–190	4Bbottom			3700 ± 50		

but the vertical distance between the 3A and 4A horizons of the paleopodzols was 20 cm less in the 2002 profile. Because the two profiles were not identical, control samples were taken for ^{14}C dating of the humic acid fractions.

OSL dating on quartz grains was performed in the luminescence dating laboratory at Ghent University. The methodology, luminescence characteristics of the samples and OSL dating results have previously

been presented by Vandenberghe et al. (2005). General information on the dating procedures and techniques as used in the Ghent laboratory can be found in Vandenberghe (2004) and Vandenberghe et al. (2004, 2009). In the following, the most relevant experimental details of the analyses are summarized.

The samples were taken by hammering stainless steel cylinders into freshly cleaned exposures. Separate samples were collected for

Fig. 2. Pollen diagram of profile Defensiedijk 1984.

evaluation of the time-averaged moisture content. In the laboratory, quartz grains from the 90–125 μm fraction were extracted from inner cores of the sampling tubes using conventional sample preparation techniques (HCl , H_2O_2 , sieving, heavy liquids, and HF). The purity of the quartz extracts was confirmed by the absence of a significant infrared stimulated luminescence (IRSL) response at 60 °C to a large regenerative β -dose. The sensitivity to infrared stimulation was defined as significant if the resulting signal amounted to more than 10% of the corresponding blue light stimulated luminescence (BLSL) signal (Vandenbergh, 2004).

Luminescence measurements were performed using an automated Risø-TL/OSL-DA-15 reader, equipped with blue ($470 \pm 30 \text{ nm}$) LEDs and IR (875 nm) diodes. All luminescence emissions were detected through a 7.5 mm thick Hoya U-340 UV filter. Details on the measurement apparatus can be found in Bøtter-Jensen et al. (2003).

The equivalent dose (D_e) was determined using the single-aliquot regenerative-dose (SAR) protocol (Murray and Wintle, 2000). Optical stimulation with the blue LEDs was for 40 s at 125 °C; the initial 0.32 s of the decay curve was used in the calculations, less a background derived from the last 4 s of stimulation. The effect of preheating on the D_e was investigated. For the youngest samples, the D_e is independent of preheat temperatures in the range of 160 °C to 200 °C. This plateau extends to higher temperatures as samples get older; the D_e in the oldest samples is insensitive to preheat temperatures up to 280 °C. Across the plateau region, all samples behaved well in the SAR protocol, with recycling ratios falling within the 1.0 ± 0.1 range and growth curves passing close to the origin. The suitability of the SAR measurement conditions was confirmed through a dose recovery test (Murray and Wintle, 2003); the given dose could be recovered to within 5%.

Radionuclide activities were measured using low-level gamma-ray spectrometry in the laboratory, and converted to dose rates using the factors tabulated by Adamiec and Aitken (1998). The external beta dose rate was corrected for the effect of attenuation and etching following Mejdahl (1979). Both the beta and gamma contributions were corrected for the effect of moisture, assuming a time-average water content of $10 \pm 3\%$. The contribution from cosmic radiation was calculated following Prescott and Hutton (1994). Based on Vandenbergh et al. (2008), an internal dose rate of $0.010 \pm 0.002 \text{ Gy/ka}$ was adopted.

3. Results and discussion

3.1. Pollen analysis

Pollen diagram Defensiedijk-1 (Fig. 2) shows a record of 4 cycles in landscape development. Cycle 1 starts with the deposition of Late Weichselian coversand (formation 1), followed by the Holocene development of a carbic podzol. The vertical distribution of pollen concentrations of zone 1S indicates post-sedimentary pollen infiltration in coversand. The pollen spectra show decreasing percentages of deciduous trees, mainly *Corylus* and *Alnus*. The percentages of Ericaceae are increasing. The radiocarbon age of HAC fraction of the 4Ah horizon indicates that around 3615 BP the forest had been degraded already into heath.

Cycle 2 starts with deposition of the Pre-Medieval driftsand (formation 2), followed by the development of a carbic podzol. Zone 2D shows low (sin-sedimentary) pollen concentrations. There is a slight increase of Gramineae, indicating some degradation of heath in the surrounding. Also the *Pinus* percentages are relatively high. *Pinus* is before 1500 AD not present in the region, but the influx of *Pinus* pollen, due to long distance transport, results in relatively high percentages in the sin-sedimentary pollen spectra. Zone 2S shows the vertical distribution of post-sedimentary pollen infiltration, dominated by Ericaceae.

Fig. 3. 2A horizon. Post-sedimentary intertext distributed organic aggregates with the intern fabric of fecal pellets; soil formation as the result of litter decomposition by fungi and micro arthropods.

Cycle 3 starts with the deposition of Medieval driftsand (formation 3). The sin-sedimentary pollen concentrations of the beds 2C3, 2C4, 2C5, 2C6 and 2C7 ($\log D > 4$) indicate a relatively low sedimentation rate. Pollen spectra are dominated by Ericaceae. Gramineae are increasing, pointing to some degradation of the heath. The pollen concentrations of the beds 2C2 and 2C1 ($\log D < 4$) indicate a higher sedimentation rate. The percentages of Ericaceae are decreasing, Gramineae increasing, pointing to serious degradation of the heath. During the next stable period (3S), a haplic arenosol (micro podzol) could develop. The 2A horizon of the micro podzol shows pollen spectra with increasing percentages of *Pinus*. Plantation of pine trees to stabilize driftsand landscapes started in The Netherlands after 1550 AD.

Cycle 4 starts with the sedimentation of the post medieval driftsand (formation 4). Since 1995 the area is stabilizing under a vegetation of grasses. Soil formation starts with the development of a rhizomull humus form.

There are no palynological indications that climatic change was responsible for periods with sand drifting. Human influence is the dominant factor.

3.2. ^{14}C dating

Table 1 summarizes the radiocarbon ages of bulk samples from 1984 and the humin and humic acid fractions of SOM, extracted from buried humic horizons, sampled in 1986 (preliminary published in Van Mourik et al., 1988) and the control samples of profile 2002. The dating results show relevant differences between the fractions and

Fig. 4. 2A horizon. Intern fabric of organic aggregates, showing the incorporation of small charcoal fragments and pollen grains.

Fig. 5. 2C horizon. Rounded, transported charcoal particles, indicating sin-sedimentary contamination by older organic matter.

depth inside the same horizon. Some interesting observations are: in the 2, 3 and 4Ah horizons, HUM seems to be 'older' than HAC. This sustains the idea that the difference in age between these fractions correlates with the duration of active soil formation. It is also clear that the age of the HAC, extracted from the upper part of a buried A horizon, will be most close to the moment of fossilization of the soil and the start of a new cycle. Another pedological interesting feature, not relevant for the chronology of driftsand deposits, is the age of SOM in the 4B horizon. The ages of HAC are similar to the 4A horizon, but in reversed order, probably pointing to a decrease of illuviation depth during active podzolation. The age of HUM (due to low concentrations, the extractions have been processed as one sample 185–190 cm) is younger than HUM in the 4A horizon. The ages of SOM fractions provide more insight in the quality of the dates for chronological interpretation. So it is clear that transported and redeposited HUM 'contaminates' SOM of the 2A horizon (sustained by soil micromorphology). But it remains problematic to correlate the fractionated ^{14}C dates with the geochronology of periods of sand drifting and soil formation in the sequence. Consequently, the measured radiocarbon ages cannot be considered as reliable for accurate absolute dating. This is also illustrated by the radiocarbon ages of the control samples of profile 2002. The ^{14}C ages of HAC, extracted from the 2A and 3A horizons, are in line with the ages obtained for the 1986 profile. The age of HAC from the 4A horizon is younger, pointing to a higher degree of rejuvenation of the original SOM; this may be caused by the shorter vertical distance to the overlying younger podzols. We have to conclude that radiocarbon ages of buried humic horizons cannot be

Fig. 7. 3A horizon. Post-sedimentary, aged intertextic distributed organic aggregates, indicating undisturbed soil structure.

used for the chronology of driftsand sequences. The complexity of SOM in such horizons is confirmed by observations in thin section.

3.3. Soil micromorphology

Micromorphological observations in thin sections of soils and sediments can improve our knowledge about the sources and complexity of SOM (Figs. 3–10). The 2A horizon is the result of post-sedimentary decomposition of leaves and roots of the vegetation during the stable

Fig. 8. 3A horizon. Channels, indicating disturbed soil structure by vertical activities.

Fig. 6. 2C horizon. Rounded, transported organic aggregate, indicating sin-sedimentary contamination by older organic matter.

Fig. 9. 4A horizon. Post-sedimentary, aged intertextural distributed organic aggregates, indicating undisturbed soil structure.

Fig. 10. 4A horizon. Channel, indicating vertical activities in the soil horizon after burying.

period of the third cycle. But in the intern fabric of the individual organic aggregates are charcoal particles visible. They have been consumed by micro arthropods together with fresh supplied litter and are responsible for 'increase' of the radiocarbon ages of bulk samples and especially the humin fraction. Sin-sedimentary charcoal particles and even organic aggregates, present in the 2C horizon explain the

contamination of driftsand with SOM, originating from eroded, older soil horizons in the environment. Fecal pellets are also the optimal micro environment for the preservation of pollen grains (Van Mourik, 2003). They are part of the fresh litter supply, but pollen grains are also incorporated in sin-sedimentary transported aggregates. That complicates the interpretation of pollen spectra, extracted from buried humic soil horizons.

Distribution pattern and intern fabric of organic aggregates in the 3A and 4A horizons are in agreement with the image of undisturbed fossilized soils. But the presence of channels points to disturbance after burying.

3.4. Pyrolysis-gas chromatography/mass spectrometry and thermally assisted hydrolysis and methylation (THM)

The pyrolysates of the HAC and HUM from horizon 2A are shown in Fig. 11. HAC shows pyrolysis products of lignin (guaiacol, 4-vinylguaiacol, 4-acetylguaiacol), polysaccharides (2-furaldehyde, 5-methyl-2-furaldehyde, levoglucosenone, levoglucosan), phenols and diketodipyrrole. In addition, a series of *n*-alkenes/*n*-alkanes (C_{10} – C_{33}) was observed. The relative abundance of this series decreases after C_{22} . The HUM fraction is dominated by these alkenes/alkanes (C_{10} – C_{33}) series. A predominant C_{31} and C_{33} alkanes were found, most likely are these from the wax layer of *Calluna*. Also a series of 2-methylketones (C_{23} – C_{33}), with an odd over even

Fig. 11. Gas chromatograms of the pyrolysates of HAC and HUM of horizon 2A. Legend: G = guaiacol; DKDP = diketodipyrrole; ▼: *n*-alkene and *n*-alkane (pair); ■: 2-methylketone; ♦: alkanic acid; C_n indicates chain length.

predominance was identified. A few pyrolysis products derived from polysaccharides and lignin were found, but only in very low abundance. Together, HAC contains still some plant derived compounds, such as lignin and polysaccharides, but the HUM fraction is mainly composed of aliphatic material. These homologous series of *n*-alkene/*n*-alkane doublets have been attributed to the non-hydrolyzable aliphatic biopolymers cutan and suberan (Nip et al., 1986;

Tegelaar et al., 1995). However, cutan seems to be limited to CAM plants only (Boom et al., 2005), which do not grow at the study area. C_{31} and C_{33} alkanes are characteristic additional wax alkanes of *Calluna*, suggesting that both leaves and stems/roots contributed to the HUM fraction. The combination of 2-methylketones and an alkene/alkane pattern is typical of *Calluna* bark/roots, pointing to suberan (Van Smeerdijk and Boon, 1987; Nierop, 1998). In addition, the

Fig. 12. Gas chromatograms of the pyrolysates of HAC of horizons 2A, 3A (top) and 4A. Legend: G = guaiacol; DKDP = diketodipyrrole; ▼: *n*-alkene and *n*-alkane (pair); ■: 2-methylketone; ◆: alkanoid; C_n indicates chain length.

alkenes/alkanes may be derived from non-biological aliphatic (geo) macromolecules which can be produced from low-molecular-weight lipids upon (oxidative) polymerization (De Leeuw, 2007). THM of HUM (data not shown) also revealed the ω -hydroxyalkanoic acids with chain lengths of C_{12} and C_{14} , and dehydroabietic acid. These compounds are typical of pine, the first of cutin (Nierop and

Verstraten, 2004) and the latter as a typical resin constituent (Simoneit et al., 1985). Altogether, both HAC and HUM consist mainly of recalcitrant plant molecules that were preserved upon decay and were not the result of illuviation.

The pyrolysates of the HAC fractions 2A, 3A and 4A horizons are shown in Fig. 12. The composition of 2A is already given above. As can

Fig. 13. Gas chromatograms of the pyrolysates of HAC of horizons 3B and 4B. Legend: ▼: *n*-alkene and *n*-alkane (pair); ■: 2-methylketone; ◆: alkanolic acid; C_n indicates chain length.

Table 2

Radionuclide activities used for dose rate evaluation, calculated dose rates, D_e values, optical ages, and random (σ_r), systematic (σ_{sys}) and total (σ_{tot}) uncertainties. The uncertainties mentioned with the D_e and dosimetry data are random. The uncertainties on the optical ages were calculated following the error assessment system proposed by Aitken and Alldred (1972) and Aitken (1976). The optical ages are expressed in ka, with 1 ka being 1000a.

Sample	Depth (cm)	Horizon	^{238}U (Bq/kg)	^{226}Ra (Bq/kg)	^{210}Pb (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)	Dose rate (Gy/ka)	D_e (Gy)	Age (ka)	σ_r (%)	σ_{sys} (%)	σ_{tot} (%)	(ka)
W49	45	1C1	6 ± 1	9 ± 2	7 ± 1	5.3 ± 0.3	159 ± 2	0.88 ± 0.02	0.072 ± 0.004	0.082	6.16	7.79	9.94	0.008
W54	55	1C2	5 ± 1	9 ± 1	7 ± 1	5.2 ± 0.2	172 ± 3	0.90 ± 0.01	0.090 ± 0.008	0.10	9.42	7.82	12.24	0.01
W48	70	1C3	3 ± 1	6 ± 2	3 ± 1	4.3 ± 0.2	171 ± 3	0.81 ± 0.01	0.075 ± 0.005	0.09	7.03	7.83	10.52	0.01
W44	105	2Ah	5 ± 1	5 ± 1	4 ± 1	5.3 ± 0.2	150 ± 3	0.78 ± 0.02	0.276 ± 0.004	0.35	2.65	7.86	8.29	0.03
W3	105	2C1	6 ± 1	10 ± 1	7 ± 1	5.4 ± 0.2	117 ± 3	0.73 ± 0.02	0.43 ± 0.01	0.59	3.22	7.80	8.44	0.05
W46	125	2C1	5 ± 1	10 ± 1	7 ± 1	5.2 ± 0.2	110 ± 2	0.69 ± 0.01	0.47 ± 0.01	0.67	3.46	7.79	8.52	0.06
W14	137.5	2C2	3 ± 1	9 ± 1	7 ± 1	4.9 ± 0.2	70 ± 2	0.57 ± 0.01	0.73 ± 0.01	1.3	3.12	7.78	8.39	0.1
W53	145	3Ah	3 ± 1	6 ± 2	4 ± 1	3.0 ± 0.2	98 ± 2	0.58 ± 0.02	3.3 ± 0.1	5.8	4.19	7.84	8.89	0.5
W21	152.5	3E	4 ± 1	7 ± 1	6 ± 1	4.1 ± 0.2	117 ± 2	0.68 ± 0.01	3.17 ± 0.02	4.7	2.39	7.81	8.17	0.4
W24	162.5	4Bh	6 ± 1	11 ± 1	7 ± 1	5.3 ± 0.2	111 ± 2	0.70 ± 0.02	6.4 ± 0.2	9.2	3.88	7.79	8.70	0.8

be seen from Fig. 12, both 3At and 4A are dominated by the *n*-alkene/*n*-alkane series, and the 2-methylketone series. With depth, the latter series increases in abundance with respect to the alkene/alkane series. Pyrolysis products of lignin and polysaccharides were virtually absent from these two horizons. Only aromatic products benzene, toluene, dimethylbenzenes and styrene were abundant in the pyrolysates of the HAC fractions, while in the HUM fraction they were hardly present (data not shown). The great similarity between the pyrolysates of 3At and 4A horizons suggests that with time (from 1365 years BP up to 4000 years BP) (Van Mourik et al., 1995), the aliphatic compounds, most likely derived from suberin of *Calluna*, or formed from low-molecular-weight lipids have accumulated. Free lipids, such as the C_{31} and C_{33} alkanes, decrease in concentration with depth suggesting that these compounds are also subject to degradation. The HAC composition indicates that the ^{14}C dates are based on the most resistant and, therefore, the oldest OM fractions.

Fig. 13 displays the GC traces of the pyrolysates of the HAC fractions of 3B and 4B horizons (top and bottom). Again, these pyrolysates are dominated by the alkene/alkane series, suggesting that with time, the illuviation horizons are dominated by compounds that are supposed to be insoluble. Typical compounds that would expect to be water soluble and candidates to be precipitated in B horizons, such as lignin-derived phenols (e.g. Nierop and Buurman, 1999), were not identified. Most likely, these compounds were degraded, and only the aliphatic compounds survived (partly) this degradation. Also, the contribution of root-derived material may have been more important than illuviation as shown by Buurman and Jongmans (2005). The HAC fractions, and particularly the HUM fractions (data not shown), provide strong indications of *Calluna* remnants, mainly in the form of roots.

Such aliphatic patterns have been observed earlier in fossil podzols B horizons in Belgium (Buurman et al., 1999) and were considered as a possible origin of aliphatic constituents in soils (Tegelaar et al.,

1989) in which even ester-bound moieties such as those derived from suberin can survive (bio)chemical degradation (Quénéa et al., 2005).

3.5. Optically stimulated luminescence (OSL) dating

Table 2 summarizes the information relevant to the age calculation, and shows the final optical dates. It can be seen that, for all but one sample (sample W54), the systematic uncertainty is dominant in the overall uncertainty on the ages, which varies in between ~8% and 12% (1σ). For the younger samples, the precision is limited by the low intensity of the luminescence signals; this accounts for random uncertainties in the range of 6%–9%, compared to 2%–4% for the older samples.

Within analytical uncertainty, the ages for the uppermost seven samples (samples W49 to W14) are consistent with the stratigraphic position of the samples. The two following samples (sample W53 from the 3Ah horizon and sample W21 from the 3E horizon) show an apparent age inversion. Field observations point to complicated sedimentary structures (Fig. 14), indicating short distance re-sedimentation processes; such reworking may explain the observed age reversal. Sample W24 was collected from the coversand unit at the base of the profile; it yields an age of 9.2 ± 0.8 ka. While this age is not stratigraphically inconsistent, it must be considered as too young. Indeed, coversand deposition is generally assumed to have stopped in the Late Glacial (Kasse, 2002). Micromorphological observations in thin sections and field observations (Fig. 15) point to some bioturbation (channels), responsible for vertical transport of organic matter and mineral grains. That means that the mineral environment in the coversand deposit can be contaminated with transported grains from the oldest driftsand deposit and reversed, causing underestimating of

Fig. 14. Detail of the complex sedimentary structure of the driftsand deposits on the 3Ah horizon (profile 1984).

Fig. 15. Detail of the deepest podzol with evidence of (paleo) bioturbation through the 3Ah horizon (profile 1984).

Table 3
OSL based geochronology of profile Defensiedijk.

Cycle	Sedimentation OSL ages (a)	Soil formation OSL ages (a)
4 Driftsand	100–recent	Initial
3 Driftsand/micropodzols	1200–0350	350–150
2 Driftsand/podzols	5000–4500	4500–1200
1 Coversand/podzols	>–9200	9200–5000

the OSL age of the topsoil in coversand and overestimating of the OSL age of the oldest driftsand deposit.

It has been demonstrated that bioturbation may have a significant effect on a luminescence age and that it not necessarily leads to OSL ages that are stratigraphically inconsistent (Bateman et al., 2003, 2007; Vandenberghe et al., 2009). As such, it cannot be excluded that more samples, or even the entire profile, are affected by post-depositional mixing to some extent. This remains to be further investigated and would require that, for each sample, the dose distribution is measured in small aliquots, which are composed of only a few grains, or even single grains of quartz.

Based on the OSL dating results (Table 2), we have a now better impression of the chronology of the polycyclic sequence (Table 3). The ages allow distinguishing multiple phases of driftsand formation within the past ~1.3 ka ago, and point at an additional period of landscape instability around 5 ka ago.

4. Conclusions

For the interpretation of paleoecological information, derived from polycyclic records, it is relevant to use soil micromorphology. SOM in the buried A horizon of the micropodzols is contaminated with older charcoal particles, resulting in an overestimation of the ^{14}C ages.

SOM in the buried A horizons of the older podzols seems not be contaminated, but the presence of channels proves some bioturbation. The chromatograms of pyrolysates of SOM extractions point to alteration of the chemical composition (decomposed leave compounds to root compounds) after burying by younger driftsand. The result is an underestimation of the ^{14}C ages.

The pollen content of buried A horizons is part of HUM. Due to processes as pollen incorporation in excremental aggregates and (bio) infiltration into the soil, the ^{14}C ages of pollen spectra are dissimilar to the OSL age of the sediments, but the pollen profile is useful for the generation of paleoecological information.

OSL dating of quartz is a powerful tool for establishing a chronological framework for polycyclic sequences in cultural landscapes. The method allows distinguishing the instable and stable periods during an individual cycle, which is not possible through ^{14}C dating. Our results also illustrate (the limit on) the time resolution that can be achieved, and exemplifies the limitations imposed by post-depositional mixing and reworking on the accuracy and precision of OSL dating in this type of sedimentary environment.

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