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Short communication

Archaeological cereals as an isotope record of long-term soil health and anthropogenic amendment in southern Scandinavia

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ABSTRACT

Maintaining soil health is integral to agricultural production, and the archaeological record contains multiple lines of palaeoclimatic and palaeoenvironmental proxy evidence that can contribute to the understanding and analysis of long-term trajectories of change that are key for contextualizing 21st century global environmental challenges. Soil is a capital resource and its nutrient balance is modified by agricultural activities, making it necessary to ensure soil productivity is maintained and managed through human choices and actions. Since prehistory this has always been the case; soil is a non-renewable resource within a human lifetime. Here, we present and interpret carbon and nitrogen isotope analysis of charred cereals from southern Scandinavia. Anthropogenic effects on soils are evident from the initiation of farming 6000 years ago, as is amendment to counteract its effects. The earliest cereals were planted on pristine soils, and by the late Neolithic, agriculture extensified. By the Iron Age it was necessary to significantly amend depleted soils to maintain crop yields. We propose that these data provide a record of soil water retention, net precipitation and amendment. From the start of the Neolithic there is a concurrent decrease in both $\Delta^{13}C$ and $\delta^{15}N$, mitigated only by the replacement of soil organic content in the form of manure in the Iron Age. The cereal isotopes provide a record of trajectories of agricultural sustainability and anthropogenic adaptation for nearly the entire history of farming in the region.

Author contribution

K.J.G., M.J.C., L.S., N.H.A. and D.R.G. designed the research. M.L., N.H.A., P.S.H., M.H.A. and M.J.C. performed the archaeobotany. K.J.G., D.R.G., M.L. and M.J.C. analyzed the samples. K.J.G., M.J.C. and D.R.G. wrote the paper with input from all authors.

1. Introduction

Soil is a natural capital resource and is non-renewable over the duration of a human lifetime (Orgiazzi et al., 2018). Therefore,

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understanding its long-term health is key for contextualizing 21st century global environmental sustainability (IPCC 2019). Soil health is directly related to its organic matter content (Díaz et al., 2019) and influenced by its bulk density, pH, organic carbon, nitrogen and phosphorus content (Natural Capital Committee 2019). These features are impacted by anthropogenic land use (Environment Agency 2019) and it has posed a significant research challenge to assess how prehistoric agricultural practices impacted soil health and productivity. To do so previously has required analysis of long sequences of relict or buried palaeosols, either at a local or regional scale (e.g. Breuning-Madsen et al., 2009; 2013). However, these records provide a snap shot at low temporal resolution, while the impact of human activities on soils could accumulate over centuries and millennia in some locations. Therefore, another proxy record is needed. One promising avenue is to examine the products of agriculture represented by archaeobotanical remains; a resource that can be directly radiocarbon dated and analyzed for stable isotopes.

The application of stable carbon and nitrogen isotope analyses to charred archaeobotanical macrofossils has become an important technique in archaeological science. This is because the carbon isotopic composition of agricultural remains can reflect watering regimes, aridity, amendment, and variations in atmospheric carbon (Bol et al., 2005; Fiorentino et al., 2012; Kanstrup et al., 2011; Nitsch et al., 2017), while their nitrogen isotopic composition can reflect soil amendment, increased nitrogen cycling and aridity (Bogaard et al., 2013; Fiorentino et al., 2012; Fraser et al., 2011; Kanstrup et al., 2014; Nitsch et al., 2017). Isotopic analyses of cereal grains therefore permit the evaluation of past crop husbandry practices, including the application of manure and various watering regimes (Bogaard et al., 2013; Wallace et al., 2014), as well as recording broad-scale factors, such as changes in climate and environment (Ferrio et al., 2005; Fiorentino et al., 2015). The challenge with this record is unravelling the various influences that contribute to the isotopic signature.

In our study area (Fig. 1), it is now generally accepted that the

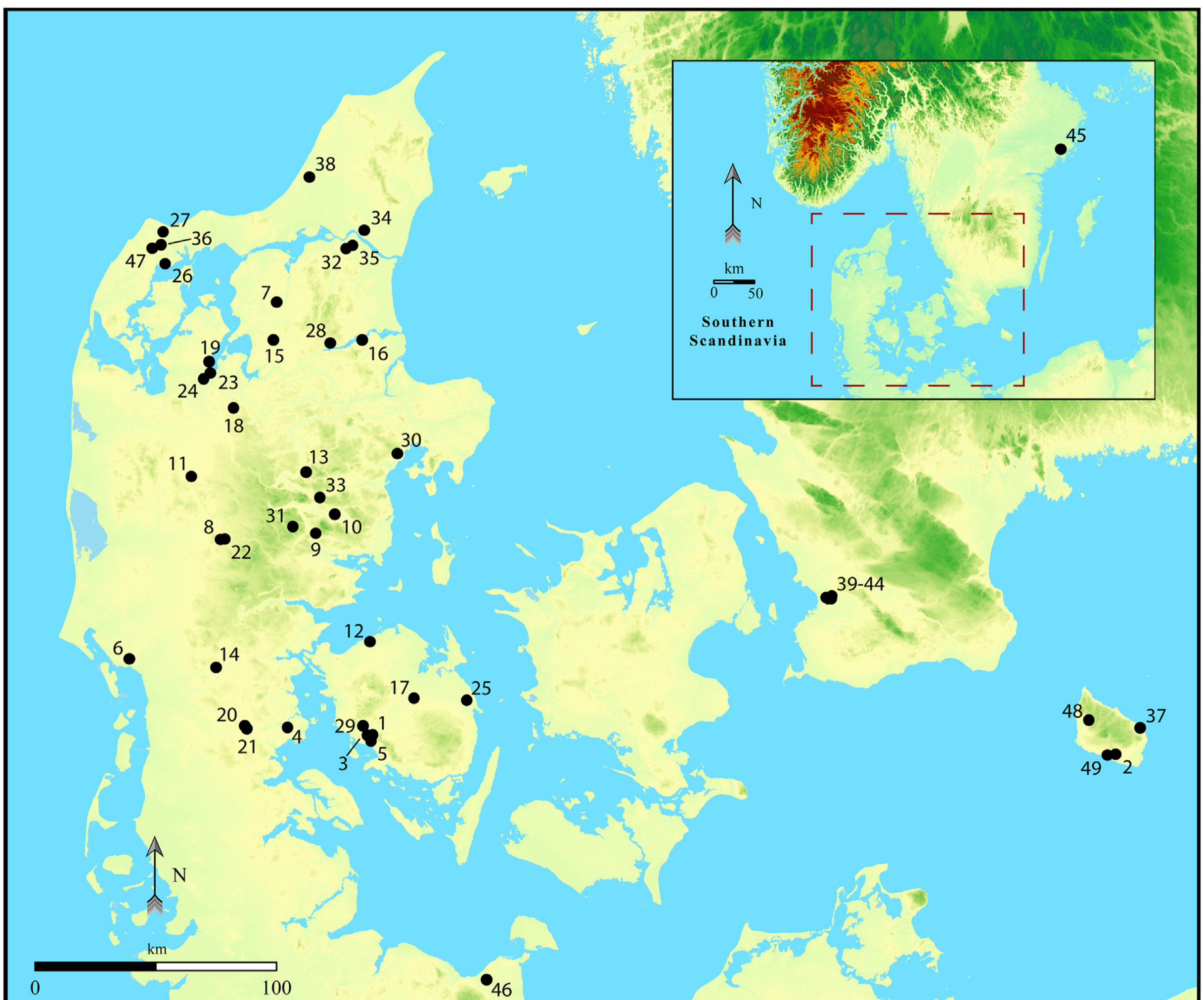

Fig. 1. Map of southern Scandinavia showing the location of archaeological sites yielding charred cereals. 1. Frydenlund. 2. Ndr. Grøbygård. 3. Sarup. 4. Lønt. 5. Damsbo Mark. 6. Uglviggård. 7. Tandrupgård. 8. Enkehøj. 9. Petersborg Vest. 10. Vrold. 11. Gilmosevej. 12. Lindebjerg. 13. Voel Boldbaner. 14. Kongehøj II. 15. Teglværksvej 31. 16. Kongsdal Huse. 17. Energivej. 18. Rosegårde. 19. Resengaard. 20. Bdr. Gram hus V. 21. Over Jernhyt. 22. Sjællandsvej V. 23. Glattrup I. 24. Kluborg II. 25. Hjulby. 26. Fårtoft. 27. Bjerre 7. 28. Bavnehøje. 29. Voldtofte. 30. Grenåvej. 31. Galgehøj. 32. Nørretranders. 33. Kildebjerg I. 34. Overbygård. 35. Nr. Hedegård. 36. Smedegård. 37. Dalshøj. 38. Blæshøj. 39. Uppåkra 8:3. 40. Uppåkra 2:14. 41. Uppåkra 12:110. 42. Uppåkra 2:25. 43. Hjärup 21:36. 44. Hjärup 9:8. 45. Stensborg. 46. Oldenburg LA 77. 47. Liselund. 48. Smedegade. 49. Limensgård. Site locations from this study, Filipović et al. 2019, Gron et al. 2017, Kanstrup et al. 2014 and Larsson et al. 2019. Basemap data © European Union, Copernicus Land Monitoring Service 2019, European Environment Agency (EEA) on which the cartography is based is from the European Digital Elevation Model (EU-DEM), version 1.1, European Environment Agency Copernicus Programme, available at https://land.copernicus.eu/imagery-in-situ/eu-dem/eu-dem-v1.1 which is produced with funding by the European Union.

expansion of the Michelsberg culture from northeastern France led to the formation of the Funnel Beaker Culture in Northern Germany, South Scandinavia and parts of Poland c. 4100–3800 BC (Sørensen 2020). Cultivating new areas meant adjusting to different environments and soil types. One of the characteristics of early Funnel Beaker agriculture in these areas was the choice to cultivate sandy soils such as those in south-west Scania (Larsson 1985), Middle Sweden (Hallgren, 2008), Bornholm (Nielsen and Nielsen 2020) and Poland (Czerniak and Rzepecki 2015). There was an obvious need for amendments of such light and potentially vulnerable soils under continuous cultivation.

In this paper, we present and interpret carbon and nitrogen isotope analysis of 327 samples of charred cereals from archaeological sites across southern Scandinavia, and interpret these together with previously published data to assess the long-term impact of agrarian practices on soil health from the Neolithic to the early Medieval period.

2. Materials and Methods

2.1. Cereal selection and recording

Ten naked barley grains (*Hordeum vulgare* var. *nudum*) and 10 durum wheat grains (*Triticum turgidum* ssp. *durum*) were selected from Frydenlund, five barley (*Hordeum* sp.) and five wheat (*Triticum* sp.) from Limensgård, 20 emmer wheat (*Triticum dicoccum*) and seven barley from Liselund, and 10 naked barley and emmer wheat from Smedegade, Denmark. This was done in order to evaluate the earliest cultivation practices in the region. Details regarding these early Neolithic sites and the associated depositional contexts can be found in Table S3. From each context, these represented the best-preserved cereals, which in most cases (82%) was Hubbard and al Azm (1990) grade P3 or better (Table S1). In some cases, the nature of the material necessitated the selection of less well-preserved grains, but our single-entity approach ensured that any systematic offset related to poor preservation could be identified and appropriately understood. Where possible, 10 charred cereals for each species were selected from each context for single-entity analysis (sensu Gron et al., 2017) to ensure comparability with bulk-sampling data derived from a minimum of 10 grains (sensu Bogaard et al., 2013): in some cases, this was not possible because not enough grains were recovered. Cereal grains were identified, selected, measured in three dimensions (X, Y, and Z, Figure S1), weighed, individually photographed and assigned preservation grade (Hubbard and al Azm, 1990), sensu Gron et al. (2017). Additionally, we include 250 Iron Age δ^{13}C values obtained in the course of previous research, but which were never published (Larsson et al., 2019). These samples are mostly barley as this cereal was the predominant cultivar in the Iron Age, and therefore dominates the available sample from southern Scandinavia (Engelmark 1992; Grabowski 2011; Larsson 2018; Robinson et al., 2009; Viklund 1998). We only include directly dated contexts from this research, in order to ensure comparability in the datasets. The archaeobotanical methods, find contexts, and analytical methods can be found in Larsson et al. (2019). Detailed data for each individual sample and information about the sites and site contexts from which they derive are listed in Tables S1, S2, and S3.

2.2. Isotopic methods

Each cereal grain was crushed to a powder and analyzed using a Costech Elemental Analyser (ECS 4010) attached to a Thermo Scientific Delta V Advantage IRMS in the Stable Isotope Biogeochemistry Laboratory (SIBL) operated by the Department of Earth Sciences, Durham University. Isotopic accuracy was actively monitored through the analyses of in-house standards (Glutamic Acid, δ^{13}C = −11.00‰, δ^{15}N = −7.50‰; IVA Urea, δ^{13}C = −43.26‰, δ^{15}N = 0.56) and was calibrated against accepted international standards (USGS40, USGS24, IAEA-600, IAEA-N-1, IAEA-N-2, NBS 19), providing linear δ^{13}C and δ^{15}N ranges for accurate corrections. Replicability error was typically ±0.1‰ (1 sd) for the international standards and <0.2‰ for sample replicates. Organic carbon and nitrogen data were obtained using an internal standard (Glutamic Acid, 40.82% C, 9.52% N) during the isotopic analysis.

Additionally, we report carbon isotope data (δ^{13}C) obtained in the course of the research undertaken by Larsson et al. (2019) which are presented here for the first time as well as the δ^{15}N values uncorrected for charring to ensure comparability with the other data. Comprehensive methods are reported in Larsson et al. (2019). Data were calibrated against accepted international standards (USGS40 and USGS41a). Replicability error (1 sd) averaged ±0.1‰ for the international standards and <0.2‰ for sample analysis with a maximum error of 0.8‰ for carbon sample analysis.

Long-term δ^{13}C data were normalised relative to variation in atmospheric CO_2 (Eggleston et al., 2016) through the method of Ferrio et al. (2005) in order to calculate Δ^{13}C.

2.3. Comparative methods

Any systematic bias stemming from variation in preservation in our dataset was first discounted through comparison of mean δ^{13}C (one-way ANOVA ($F(2,72) = 1.75894$, $p = 0.18$)) and δ^{15}N (one-way ANOVA ($F(2,72) = 1.12165$, $p = 0.33$)) values of cereals with Hubbard and al Azm (1990) preservation grades 2, 3, and 4. We apply no correction to the isotope ratio measurements for charring (Fraser et al., 2013; Nitsch et al., 2015) because the temperature and atmospheric conditions of charring are unknown. Nonetheless, even if a very conservative correction of 1‰ (Fraser et al., 2013), larger than the more common 0.3‰ correction (Nitsch et al., 2015), is applied to the average δ^{15}N values, our interpretations remain unchanged. Overall variance is similar between wheat and barley, suggesting no systematic difference originating from variability in preservation condition and by extension charring temperature and duration (Styring et al., 2013). A similar comparison with the Kanstrup et al. (2014) and Larsson et al. (2019) datasets was not possible due to a lack of systematic recording of preservation grade for each individual cereal grain. At Stensborg, no systematic bias relating to preservation was identified between preservation grades 2 and 3 (Gron et al., 2017). All LA77 (Filipović et al., 2019) cereals are reported as Hubbard and al Azm (1990) preservation grade 3, but previous research (Gron et al., 2017), and the above results, suggest that there is no reason to suspect any systematic offset in comparisons of isotope measurements on cereals of this grade with those of other grades. Given the nature of the available dataset, bulk and single-entity isotope measurements were selected for comparison. Single-entity values by context from our new data, and from previous studies (Gron et al., 2017; Larsson et al., 2019) were averaged in order to be compared with bulk data from Filipović et al. (2019) and the non-pretreated cereals from Kanstrup et al. (2014). Radiocarbon ages were recalibrated using OxCal 4.3, IntCal 13 (Reimer et al., 2013; Table S2) and displayed as the midpoint of the 2σ range (Fig. 2). Whilst a comprehensive dating programme has been undertaken at Frydenlund (Andersen 2019), individual cereal context dates are used here to ensure comparability with the wider dataset. The data from Filipovic et al. (2019) were averaged to be a single data point for barley and a single data point for wheat and are displayed on Fig. 2. The date given is the midpoint for the date range given in Filipovic et al. (2019).

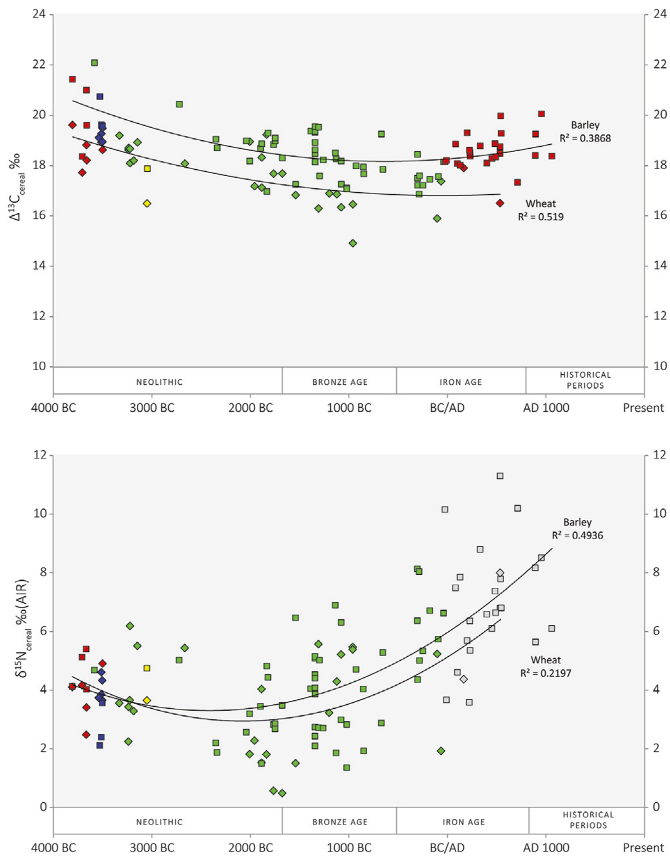

Fig. 2. Charred cereal $\Delta^{13}C$ (top) and $\delta^{15}N$ (bottom) values representing five thousand years of Scandinavian prehistory. Bulk data, or mean single-entity values per context shown. Squares represent barley and diamonds represent wheat. Red values (this study), blue values (Gron et al. 2017), yellow values (Filipović et al. 2019), green values (Kanstrup et al. 2014), and grey values (Larsson et al. 2019). Iron Age $\delta^{13}C$ data used to calculate $\Delta^{13}C$ in red was obtained by Larsson et al. (2019) but not published. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Lastly, in order to investigate diachronic change in grain size, estimates of pre-charring cereal size were applied where three-dimensional measurements of individual cereals were available (e.g. this study; Gron et al., 2017; Larsson et al., 2019). Following the regression equations given in Ferrio et al. (2004), estimated pre-charring grain weight was calculated for length versus width (Figure S1, here X vs Y; Table S1; Ferrio et al., 2004) and length versus thickness (Figure S1, here X vs Z; Table S1) in our new data. The same was applied to the previously published cereals from Stensborg (Table S1; Gron et al., 2017).

3. Results and discussion

New single-entity carbon and nitrogen isotope values are presented for 77 charred cereals dating to the early Neolithic Funnel Beaker Culture (EN TRB, 4000-3300 cal BC) of southern Scandinavia. Summary results and associated statistics are listed in Table 1.

In order to contextualize our data in the long-term, we also include the cereal data from Kanstrup et al. (2014), with recalibrated ^{14}C dates according to a more recent calibration curve (Reimer et al., 2013). We then consider these individual grain measurements alongside averaged values of multiple bulk samples from Filipovic et al. (2019), Gron et al. (2017) and Larsson et al. (2019) (Fig. 2; site locations in Fig. 1; data in Tables S1 and S2). Larsson et al. (2019) reported only $\delta^{15}N$ data. Therefore, we present their $\delta^{13}C$ data for 250 samples in this study (Table S1; Table 1; Materials and Methods). All $\delta^{13}C$ data were normalised relative to fluctuations in atmospheric CO_2 ($\Delta^{13}C$) to ensure comparability over the long-term (Materials and Methods).

Manuring of cereals has been determined experimentally by $\delta^{15}N$ values higher than 3‰ (Bogaard et al., 2013; Fraser et al., 2011) due to the incorporation of recycled nitrogen into plant tissues. Our new data demonstrate that the majority of the early Neolithic cereals are in the manured range, with a few exceptions. This suggests that manuring was practised from the very start of agriculture in southern Scandinavia (Fig. 2). When taken in context with previously published data from the Later Neolithic and Bronze Age, there is greater variability in observed $\delta^{15}N$ values, probably indicating extensification and differential access to manure predicated on distance of fields from settlements where livestock were kept. In the Iron Age there is a noticeable rise in barley $\delta^{15}N$ values ($R^2 = 0.49$), such that all cereals (with one exception, a wheat value) fall into the manured range, and many into the “high” manured range (>6‰) (Bogaard et al., 2013). This would be consistent with an intensification of agricultural soil amendment in this period. The same is reflected by our new data regarding the size of the cereals themselves (Fig. 3), wherein the averaged estimated X vs Y and X vs Z pre-charring weights of the averaged individual barley grains by context are significantly higher in the Iron Age than they are even in the Early Neolithic (Supplementary Table S4; Fig. 3; $t(28) = -3.3$, $p < 0.01$). Despite an apparently similar rise in wheat $\delta^{15}N$ values (Fig. 2; $R^2 = 0.22$), the lack of wheat data from this period means it is unclear if this is an artefact of small sample size, and therefore this result should be treated with caution until additional data are available.

Cereal $\Delta^{13}C$ in the compiled data set (this study; Kanstrup et al., 2014; Filipović et al., 2019; Gron et al., 2017; Fig. 2) range between ca.15‰ and 22‰. Both wheat and barley show a consistent decrease until the end of the Bronze Age, after which a rise is documented ($R^2 = 0.39$ and $R^2 = 0.52$ respectively) (Fig. 2). This trend does not differ between the species (two-tailed Fisher R to Z, $p = 0.327$) and barley average $\Delta^{13}C$ values are consistently ~1‰ higher than wheat; an offset previously observed by Wallace et al. (2014). From the Neolithic to the Bronze Age, barley $\Delta^{13}C$ values fall from ca. 20.5‰–18.5‰, and wheat from ca. 19‰–17‰. In conjunction with this, barley $\Delta^{13}C$ values rise during the Iron Age to early Medieval Period, but never attain maximum values as high as those from crops grown on the pristine soils of the earliest Neolithic.

Plant $\Delta^{13}C$ is mainly controlled by stomatal conductance and photosynthetic activity. Other factors such as water stress (Ferrio et al., 2005), mean annual precipitation (Diefendorf et al., 2010), c_i/c_a ratios, photosynthetic activity and pCO_2 levels (Polley et al., 1993; Zhang et al., 2019) can also be contributory. Globally, there is a decrease in net precipitation at mid-latitudes through the Holocene (Routson et al., 2019). However, individual proxy records in Denmark indicate precipitation remained either relatively constant (Brown et al., 2011) or fluctuated between wet and dry periods (De Jong et al., 2009; Olsen et al., 2010). The large number of sites represented by our study likely encompasses the range of variation in local conditions, and therefore reflects regional environmental conditions and trends through time. It is difficult to explain the patterns in the isotopic data by invoking shifts in precipitation and water stress that change $\Delta^{13}C$ values.

Pre-Industrial carbon dioxide concentrations have increased from about 260 ppmV to 280 ppmV since the early Neolithic (Elsig et al., 2009; Indermühle et al., 1999), which would subsequently affect the c_i/c_a ratio in plants and hence, isotope fractionation

Table 1
Summary data obtained in this study. Italicised data previously published in Larsson et al. (2019).

Archaeological Site	Sample ID	Find Context	Period	Radiocarbon age (BP)	(overall 2σ range) Oxcal 4.3, IntCal 13	Number of Grains	Cereal type	Range, Mean, SD $\delta^{13}C$ (‰)	Range, Mean, SD δ^{15N} (‰)
Limensgård	LG IS.1-5	FJ3, FJ4, FJ5	Neolithic	5000 ± 70	3950 to 3660 BC	5	Barley	−27.86 to −25.40, −27.22 ± 1.02	0.43 to 7.39, 4.13 ± 2.95
Liselund	LIS IS.41-47	N40	Neolithic	5082 ± 29 to 4643 ± 29	3961 to 3361 BC	7	Barley	−27.47 to −23.74, −25.47 ± 1.22	1.83 to 5.86, 4.03 ± 1.42
Smedegade	SM IS.1-10	A88	Neolithic	4917 ± 32	3766 to 3645 BC	10	Barley	−25.78 to −23.39, −24.28 ± 0.67	1.85 to 7.39, 5.13 ± 1.71
Frydenlund	FRY IS.1-10	A36, A89, A106	Neolithic	4936 ± 67, 4756 ± 28, 4756 ± 28, 4807 ± 38, 4948 ± 29, 4771 ± 32	3943 to 3384 BC	10	Barley	−27.62 to −25.54, −26.81 ± 0.64	2.71 to 13.77, 5.40 ± 3.48
Liselund	LIS IS.11-20	N53	Neolithic	4688 ± 49	3631 to 3366 BC	10	Wheat	−25.47 to −23.66, −24.53 ± 0.59	1.76 to 10.67, 4.91 ± 2.30
Liselund	LIS IS.31-40	N40	Neolithic	5082 ± 29 to 4643 ± 29	3961 to 3361 BC	10	Wheat	−25.12 to −23.37, −24.13 ± 0.47	1.30 to 7.06, 3.41 ± 1.52
Smedegade	SM IS.11-20	A88	Neolithic	4917 ± 32	3766 to 3645 BC	10	Wheat	−25.26 to −22.06, −23.67 ± 1.02	2.14 to 7.40, 4.17 ± 1.51
Limensgård	LG IS.6-10	FJ5	Neolithic	5000 ± 70	3950 to 3660 BC	5	Wheat	−26.12 to −24.97, −25.49 ± 0.52	2.99 to 5.38, 4.10 ± 0.90
Frydenlund	FRY IS.11-20	A36, A147	Neolithic	4936 ± 67, 4788 ± 38, 4801 ± 29, 4880 ± 44, 4771 ± 31	3943 to 3384 BC	10	Wheat	−25.42 to −23.33, −24.72 ± 0.63	1.40 to 5.10, 2.48 ± 1.09
Regional centre Uppåkra		Hall-building/Profile 87033	Iron Age	1990 ± 50, 2035 ± 50	175 BC to 126 AD	10	Barley	−26.27 to −23.15, −24.11 ± 1.01	*3.36 to 18.58, 10.16 ± 4.16*
Regional centre Uppåkra		House/Profile 110342	Iron Age	1915 ± 45	20 BC to 223 AD	10	Barley	−25.29 to −22.73, −24.03 ± 2.23	*0.60 to 8.38, 4.61 ± 2.23*
Regional centre Uppåkra		Trench B	Iron Age	1935 ± 45	44 BC to 210 AD	10	Barley	−25.93 to −23.50, −24.77 ± 0.73	*4.80 to 9.89, 7.49 ± 1.68*
Regional centre Uppåkra		Hall-building/Profile 92518	Iron Age	1830 ± 50	71 to 330 AD	10	Barley	−25.85 to −24.79, −25.18 ± 0.34	*3.81 to 7.98, 5.69 ± 1.07*
Regional centre Uppåkra		House/Profile 110342	Iron Age	1890 ± 45	24 to 235 AD	10	Barley	−25.30 to −22.10, −23.96 ± 0.91	*5.07 to 10.37, 7.85 ± 1.56*
Regional centre Uppåkra		Hall-building/Profile 105776	Iron Age	1485 ± 45	430 to 651 AD	10	Barley	−26.70 to −25.39, −25.77 ± 0.35	*4.39 to 9.43, 7.79 ± 1.79*
Regional centre Uppåkra		Profile 110342	Iron Age	1505 ± 45	428 to 641 AD	10	Barley	−25.79 to −23.39, −24.61 ± 0.72	*5.71 to 18.56, 11.30 ± 3.50*
Regional centre Uppåkra		Vifots house	Iron Age	1640 ± 50	258 to 543 AD	10	Barley	−25.23 to −22.76, −24.01 ± 0.74	*5.25 to 7.20, 6.59 ± 0.57*
Regional centre Uppåkra		Bårhuset	Iron Age	1605 ± 45	346 to 559 AD	10	Barley	−24.97 to −23.52, −24.18 ± 0.46	*4.49 to 9.64, 6.10 ± 1.43*
Regional centre Uppåkra		House 11	Iron Age	1505 ± 60	425 to 645 AD	10	Wheat	−23.70 to −21.26, −22.45 ± 0.77	*6.36 to 9.63, 8.00 ± 0.87*
Regional centre Uppåkra		House 11	Iron Age	1505 ± 60	425 to 645 AD	10	Barley	−25.93 to −23.32, −24.35 ± 0.76	*4.36 to 11.53, 6.80 ± 2.27*
Regional centre Uppåkra		Hall-building/House 24	Iron Age	1580 ± 45	390 to 577 AD	10	Barley	−26.92 to −22.76, −24.74 ± 1.22	*5.22 to 11.08, 7.38 ± 2.05*
Regional centre Uppåkra		Hall-building/House 23	Iron Age	1485 ± 50	329 to 652 AD	10	Barley	−25.14 to −23.03, −24.22 ± 0.61	*5.02 to 10.79, 6.65 ± 1.53*
Regional centre Uppåkra		Oven area	Iron Age	1310 ± 30	656 to 769 AD	10	Barley	−24.52 to −22.58, −23.23 ± 0.57	*5.98 to 14.00, 10.20 ± 0.57*
Regional centre Uppåkra		Hall-building/House 22	Iron Age/Early Medieval	1130 ± 50	773 to 1011 AD	10	Barley	−25.61 to −22.77, −24.33 ± 0.85	*4.79 to 13.99, 8.17 ± 2.57*
Regional centre Uppåkra		Ceremonial building	Iron Age/Early Medieval	1118 ± 30	778 to 1011 AD	10	Barley	−25.88 to −24.09, −25.15 ± 0.56	*2.89 to 7.90, 5.64 ± 1.31*
Uppåkra 2:14		Pithouse	Iron Age	1996 ± 39	104 BC to 84 AD	10	Barley	−25.35 to −23.36, −24.17 ± 0.61	*2.16 to 6.00, 3.67 ± 0.95*
Uppåkra 12:110		House 10	Iron Age	1836 ± 30	86 to 245 AD	10	Wheat	−25.01 to −23.54, −23.85 ± 0.52	*3.19 to 5.16, 4.35 ± 0.52*

(*continued on next page*)

Table 1 (*continued*)

Archaeological Site	Sample ID	Find Context	Period	Radiocarbon age (BP)	(overall 2σ range) Oxcal 4.3, IntCal 13	Number of Grains	Cereal type	Range, Mean, SD δ^{13}C (‰)	Range, Mean, SD δ^{15}N (‰)
Uppåkra 2:25		House 17	Iron Age	1794 ± 30	132 to 329 AD	10	Barley	−25.10 to −23.54, −24.29 ± 0.52	*3.79 to 6.47, 5.36 ± 0.84*
Uppåkra 2:25		House 31	Iron Age	1798 ± 30	131 to 326 AD	10	Barley	−24.35 to −23.60, −24.43 ± 0.47	*3.32 to 7.89, 6.36 ± 1.35*
Uppåkra 2:25		Pit	Iron Age	1817 ± 30	125 to 322 AD	10	Barley	−25.74 to −23.43, −24.52 ± 0.76	*1.40 to 6.25, 3.59 ± 1.63*
Uppåkra 2:25		Hus 4	Iron Age	1696 ± 30	255 to 411 AD	10	Barley	−26.47 to −23.43, −24.66 ± 1.04	*7.34 to 9.50, 8.80 ± 0.73*
Hjärup 21:36		Pit	Iron Age	1460 ± 50	432 to 662 AD	10	Barley	−26.01 to −23.95, −25.11 ± 0.67	*4.75 to 7.54, 6.81 ± 0.87*
Hjärup 9:8		House 11 (outside)	Iron Age/Early Medieval	1087 ± 30	893 to 1016 AD	10	Barley	−26.39 to −24.69, −25.96 ± 0.47	*6.95 to 9.27, 8.52 ± 0.84*
Hjärup 9:8		House 13	Iron Age/Early Medieval	1016 ± 30	971 to 1149 AD	10	Barley	−26.01 to −22.9, −24.36 ± 0.82	*2.97 to 10.05, 6.10 ± 2.13*

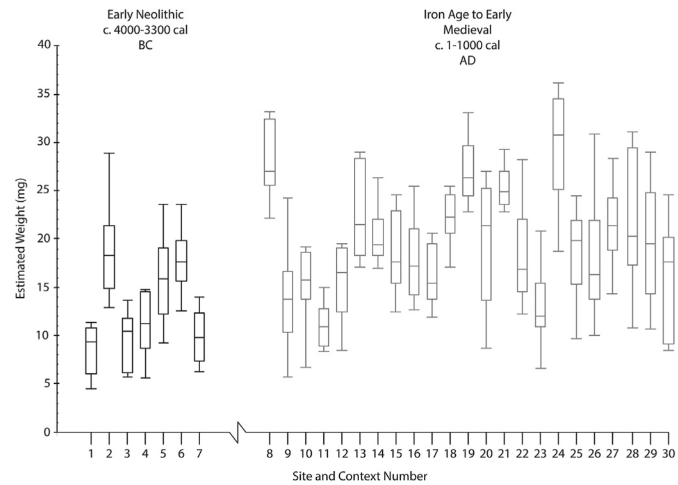

Fig. 3. Barley estimated pre-charring grain weights by context. Displayed values are an average of X vs Y and X vs Z. Data calculated in Table S1. Data are arranged in chronological order as determined by the centrum of the range of context dates (Table S2) and the individual contexts here identified numerically are detailed in Table S4.

between CO_2 and plant tissues. Although there are many studies that show increasing or decreasing Δ^{13}C with pCO_2 (see Table 1 in Zhang et al., 2019), we have chosen the study of Zhang et al. (2019) as they report a change in stomatal conductance that fits the photosynthetic fractionation model of Farquhar et al. (1989), and robustly controlled the parameters that can have an impact on carbon isotope ratios.

Recently, Zhang et al. (2019) conducted modern experiments on winter wheat (*Triticum aestivum*) under low carbon dioxide concentrations and reported that an increase in pCO_2 would result in decreased Δ^{13}C values under both watered and drought conditions. If experimental offsets of Zhang et al. (2019) are applied to our dataset, a change of 20 ppmV can only account for a change in Δ^{13}C of ca.1‰, which is approximately half of the recorded mean change in Δ^{13}C from the Neolithic to the Bronze Age (Fig. 2). Therefore, another factor must also be contributing to the long-term change in Δ^{13}C.

The organic content of a soil is a major factor that influences soil water retention; with decreased organic content, less water is retained (Hudson 1994). We propose that soil with less organic content will have lower Δ^{13}C values than a similar soil with higher organic content, due to water stress related changes in isotopic fractionation (Wallace et al., 2014). Certain aspects of cereal agriculture are known to reduce the organic content of soils (Haas et al., 1957), although the rate by which this occurs on millennial timescales is unclear.

By the Iron Age, there is evidence from δ^{15}N of an increase in the intensity of agricultural manuring. The decreasing trend in Δ^{13}C ceases at this time and in barley begins to rise. Due to very few samples, it is unclear if a concurrent rise is also seen in wheat. This cannot be attributed to a decrease in light availability owing to a reduction in openness, as the contemporary Scanian pollen record indicates the landscape remained open at the time (Berglund 1991; Lagerås and Fredh 2020). Therefore, while amendment prior to the Iron Age certainly replaced some soil organic content, this was insufficient to overcome the aggregate environmental and anthropogenic influences, causing a net Δ^{13}C reduction. However, we argue that the intensive Iron Age amendment strategy marks the point at which overall soil health starts to improve.

This strategy was a consequence of the social and economic changes that took place in the Late Bronze Age, when systems of permanent fields start to appear followed by a somewhat later aggregation of individual farms into villages or clusters of farms connected with a system of permanent fields enclosed by banks, the so-called Celtic fields. These fields were amended with household waste, animal manure and material from wetlands and heathland (Nielsen et al., 2019). Remains of Celtic fields have been found in large parts of northwestern Europe (Fries 1995). They have been documented all over southern Scandinavia (Nielsen 1993; Nielsen and Clemmensen 2010), and they were in use from the Late Bronze Age/Iron Age until c. AD 200, followed by a reorganisation of settlement and land use, leading to larger farms and new agrarian practices with an emphasis on cattle husbandry.

Within the Iron Age the period AD 200–550 saw an intensification of agricultural production. The farms grew in size, the main longhouses often having a length of more than 40 m, including room for a large byre (Jessen 2012). Hulled barley and rye were the main crops, but it is less clear how the fields were organized. However, in eastern Middle Sweden and Gotland, Celtic fields were succeeded by a system with enclosed infields and outlying common grazing areas, with the infields being intensively manured (Widgren and Pedersen 2011).

Prehistoric farmers in Scandinavia therefore understood that poorer soils could be improved through amendment. Over millennia, and in context of broader climatic changes, soil quality diminished and agriculture was diversified to a broader range of settings and soils. It is likely only by the Iron Age that the organizational structures were in-place to permit amendment on a large enough scale to revitalize soil health on a regional scale and counteract the process of agricultural soil depletion which commenced in the Early Neolithic.

Therefore, we argue that the long-term isotopic record of charred cereals from across southern Scandinavia provides information on soil health. Other potential proxies such as weed functional traits (e.g. Styring et al., 2017) do not at present rest on sufficient data to do so. Similarly, previous research directly on palaeosols provides relevant data at only a limited temporal and spatial scale, and therefore this study provides a unique record of attritional anthropogenic environmental impact on Scandinavian soils not visible through other means. This record most closely follows water availability as a function of soil organic content, which has changed over millennia as a result of agricultural practice. Broad-scale decreasing mid-latitude rainfall compounded this effect. The application of manure to enhance soil fertility and crop growth and yields was practiced in the region from the start of the Neolithic. For the first 4000 years of agriculture this practice was insufficient to maintain soil health. Intensive amendment in the Iron Age fundamentally tipped the balance in favour of large-scale widespread anthropogenic management. Therefore, the combined $\delta^{15}N$, $\delta^{13}C$, and derived $\Delta^{13}C$ data from charred cereal grains provide an integrated long-term record of human crop-husbandry strategies, precipitation, and soil conditions. This record demonstrates that climate change and environmental degradation have long presented challenges for farming populations, but these can be potentially overcome by intensive organic manuring on a regional scale.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.quascirev.2020.106762.

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