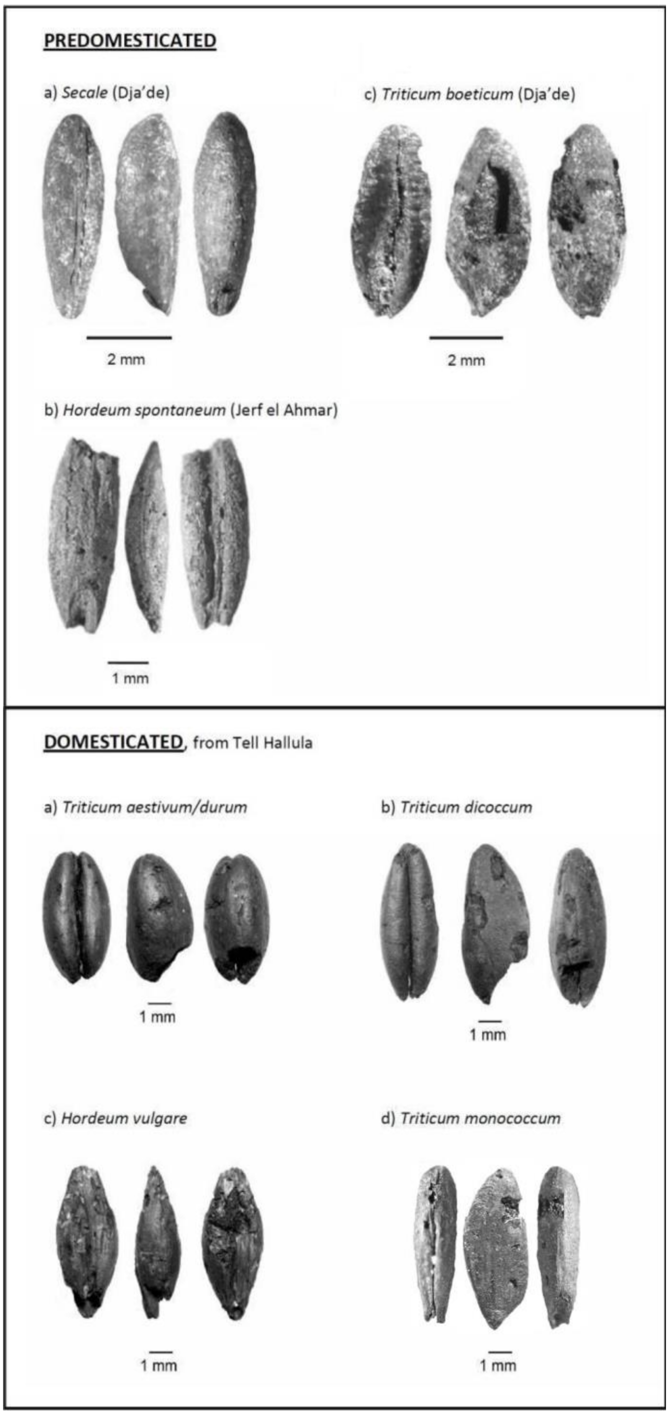
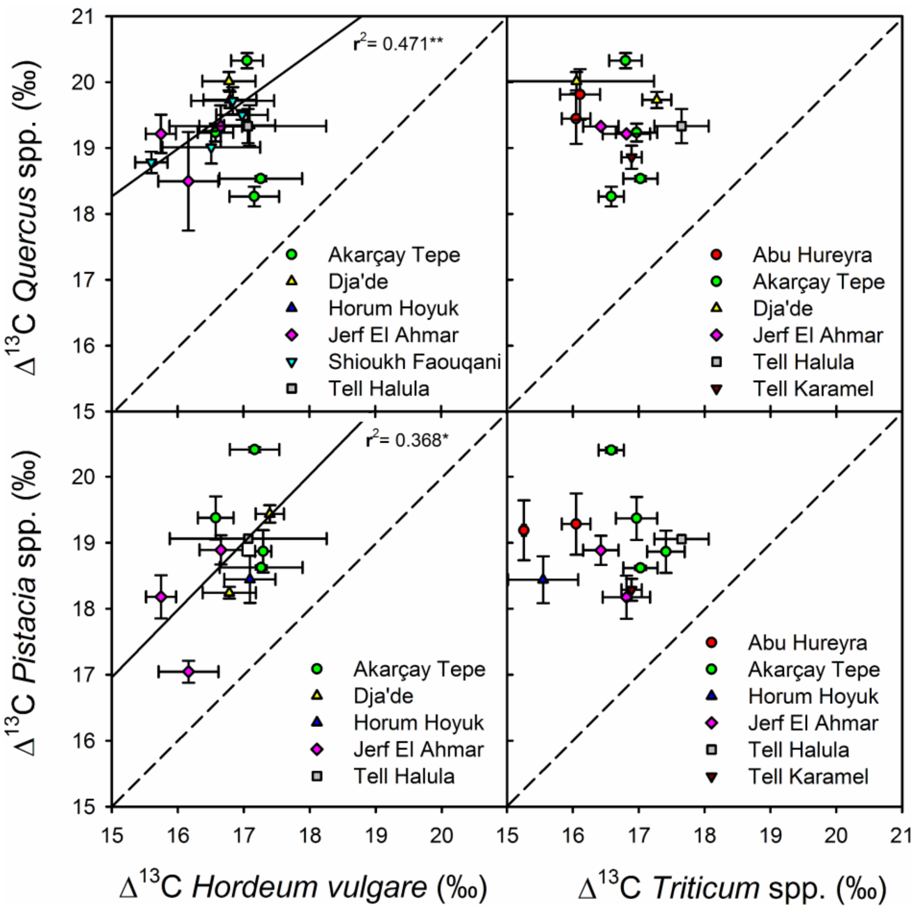
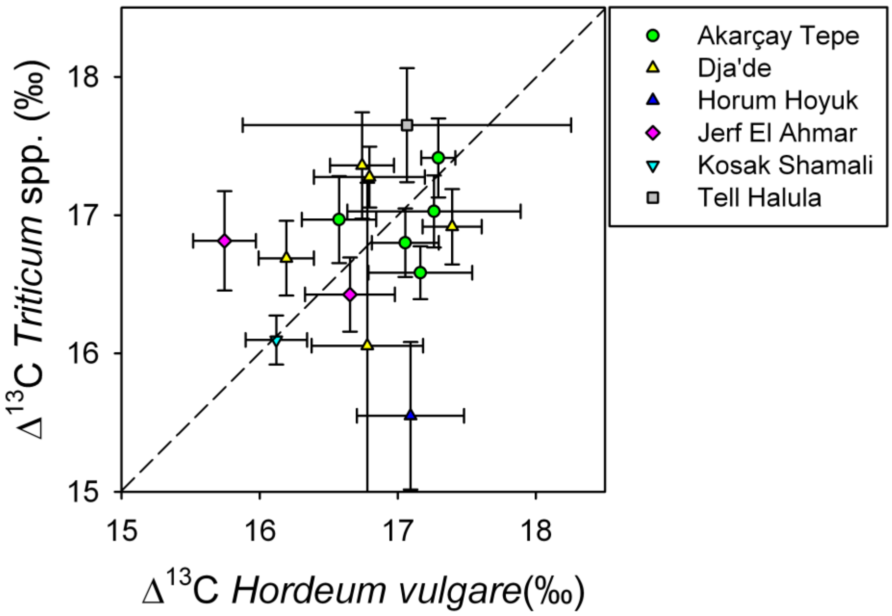
Supplementary Figure 1│Cereal species studied. The cereals studied included pre-domestication species (rye, *Secale cereale*; wild barley, *Hordeum spontaneum* and wild einkorn, *Triticum boeticum*) together with another four cereals considered as domesticated (einkorn, *Triticum monococcum*; emmer, *Triticum dicoccum*; naked wheat, *Triticum aestivum/durum* and barley, *Hordeum vulgare* subsp. *distichum*).



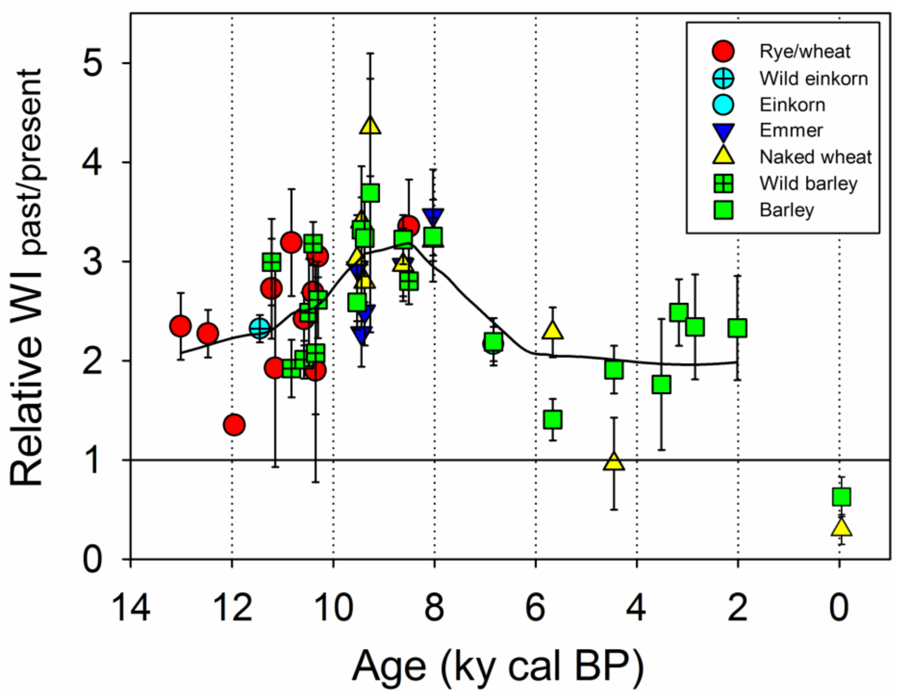
Supplementary Figure 2│Relationships between Δ^{13}C in barley (left figures) and wheat (right figures) kernels and *Quercus* (upper) and *Pistacia* (lower) charcoals. Each value represents the mean ± SE of kernels and charcoal samples from a specific site and dating. The dashed line indicates the 1:1 reference line.



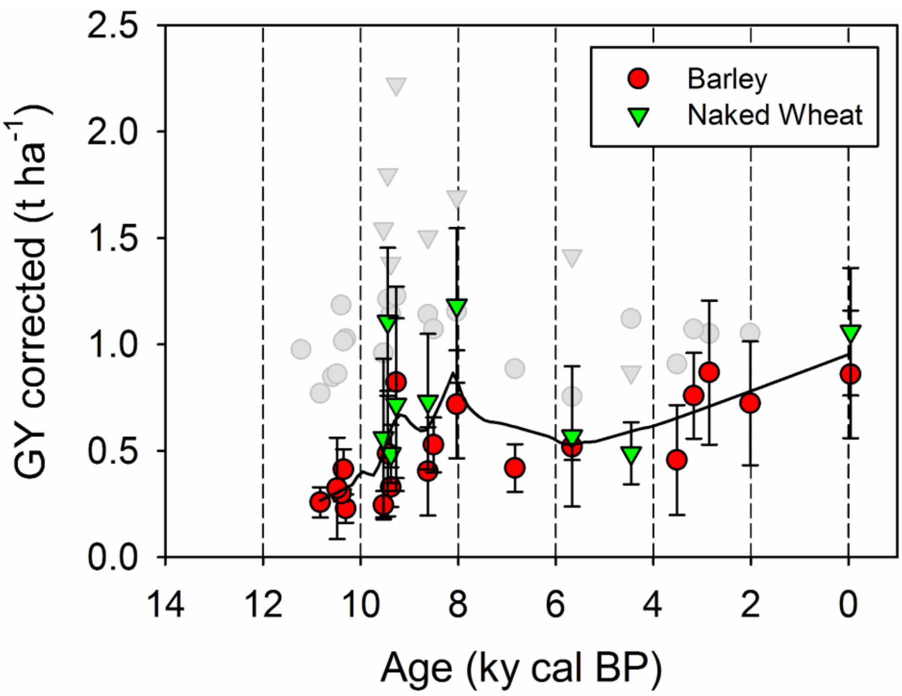
Supplementary Figure 3│Relationship between Δ^{13}C in barley and wheat kernels. Each value represents the mean ± SE of kernels samples from a specific site and dating. The dashed line indicates the 1:1 reference line.



Supplementary Figure 4│Relative water inputs during grain filling (Past/Present). The trend line depicts a locally weighted least-squares regression curve (LOESS) fitted to the data. Each value represents the mean ± SE of kernels from a particular species, site and dating. Water input in the past was estimated from $\Delta^{13}C$ values of charred kernels recovered at 11 different sites (supplementary Table 1). The horizontal line refers to the present-day average water input for rainfed barley and wheat in the region (calculated as the total precipitation accumulated during April and the first half of May).



Supplementary Figure 5│Grain yield. Evolution of grain yield (GY) of barley (*Hordeum vulgare* subsp. *distichum*) and naked wheat (*Triticum aestivum/durum*), as estimated from $\Delta^{13}C$ (grey symbols), and corrected by the lower kernel weight values in the past than in the present (red and green symbols). Each value represents the mean ± SE of kernels from a specific site and dating. Present time yields in the regions correspond to rainfed crops as detailed in the Methods section.



Supplementary Note 1: Interspecific comparison of $\Delta^{13}C$ in modern samples

To investigate species co-occurrence patterns of $\Delta^{13}C$, subsets of modern samples of different species (either forest trees or cereals) that were found to co-occur in particular geographical settings were subjected to correlation analysis involving all pair-wise species combinations. The relationship between the $\Delta^{13}C$ of *Quercus* and *Pistacia* was positive and significant (r = +0.59). Similarly, the $\Delta^{13}C$ records of *Hordeum vulgare* and *Triticum aestivum* were positively related (r = +0.97). The tighter association between the $\Delta^{13}C$ values of cereals agrees with the highly coincident temporal frames of grain filling for both species (April–May).

Supplementary Note 2: $\Delta^{13}C$ in kernels as an indicator of crop water status in the past

The $\Delta^{13}C$ values in plant remains of different crops from seven sites in northern Mesopotamia and the Levant support increased aridity from the Early Bronze Age (4700-4000 BP) to the Middle Bronze Age (4000 - 3600 BP)[1]. This trend is particularly evident in north Syria (the same area of our study) and is supported by other (more indirect) climate proxies[2]. Thus, while a relatively satisfactory water status can be recognized in the crop assemblages of Early Bronze Age sites, with the cultivation of numerous crops with comparatively high water requirements, the impact of the '4.2 ka event', with an increased aridity after 4,000 BP at the latest, is reflected strongly in the reduction or absence of drought-susceptible crop species in the Middle Bronze Age[3]. Similarly, results from the sites at Ebla and Qatna in Syria and Arslantepe in Turkey using stable carbon isotope records of plant remains also show one arid phase dating to the '4.2 ka event'[4-6].

Supplementary Note 3: Local variability in climate during the ‘4.2 ka event’

The ‘4.2 ka event’ is one of the best investigated Holocene climate fluctuations, particularly in the Near East. However, there are numerous archaeological works emphasizing the local variability in climate effects in the region (e.g. upper vs. middle Euphrates). In regard of this, there is no definite confirmation for a widespread and distinct event at 4.2 ka cal BP; rather, current evidence[7] suggests that it was mediated by the overall climatic changes recorded for the period, which commenced at approximately 4.6 ka cal BP.

Supplementary Note 4: Manuring and pre-domesticated cereals

The high $\delta^{15}N$ values obtained from 'pre-domesticated' grains may reflect some form of manuring, either as construed in the 'dump heap' theory of early cultivation[8] or in view of claims from early sites like Abu Hureyra that some of the background flora was derived from herbivore dung collected and burned as fuel[9].

Supplementary Note 5: Demography and cereal cultivation

A trend towards cereal-based farming during the Neolithic, associated with a decrease in the diversity of wild flora, has been reported for Tell Halula[10] (one of the Neolithic sites included in this study). This trend was accompanied by a growth in population during the earliest phases of the settlement (10.2 – 9.0 ka cal BP), probably enhanced by more favourable environmental conditions[2,11,12]. A decline in population followed in the late phases (9.0 – 7.4 cal BP) perhaps as a consequence of exceeding the capacity of the agroecosystem, including scarcity of cultivable land within a manageable distance from the site together with the occurrence of the '8.2 ka event'[13]. Moreover, the possible expansion of the area dedicated to agriculture would have been necessarily devoted to extensive crops requiring relatively low labour efforts, such as cereals[14,15]. In the case of Tell Halula this hypothesis is partly confirmed by a significant reduction in legume cultivation from PPNB to PN[10]. Furthermore, an expansion of cultivated area would force the use of less fertile soils as readily cultivable patches became less abundant. This may explain the reduction in cereal yields from PPNB to PN, as estimated from the carbon isotope composition of charred kernels.

Supplementary Note 6: Environmental versus evolutionary changes in seed size.

Seed size changes in domesticated cereals was in its long-term development related to an evolutionary process[16], while short-term fluctuations were most likely linked to the prevailing environmental conditions. This is in agreement with archaeobotanical evidence from northern Mesopotamian sites reporting e.g. a seed size decrease at Tell Hadidi and Tell Sabi Abyad between the Middle and Late Bronze Age[17].

Supplementary Note 7: Effect of breeding on kernel weight

Attempting to increase grain yield in breeding programs by simply choosing lines carrying heavier grains has proved mostly ineffective. For example, a complete compensation between the number of kernels and kernel weight has been reported after eight cycles of selecting heavier grain lines without any improvement in grain yield[18]. Average kernel weight is a more conservative attribute than kernel number[19-21]. Kernel weight has not been markedly modified by wheat breeding during the past century[22,23], including Mediterranean wheat[24-28], and some results have even shown that individual kernel weight was reduced by genetic improvement[29,30].

Supplementary Note 8: Climate change and social disturbances

For example, an arid event around 8,200 years ago (termed the '8.2 ka event') is sometimes linked with the abandonment of early (Pre-Pottery Neolithic) farming settlements in the Southern Levant[31,32]. In addition, a dry period that is detectable in several climate proxies around 4.2 ka cal PB (the so called '4.2 ka event') and is thought to have lasted for about 300 years has often been related to cultural collapse, for example the breakdown of the Akkadian empire[31-34]. In any case, Neolithic sites in the region had started to decline in terms of demography prior to the dry '8.2 ka event' that marked their abandonment. This has been reported for both major settlements such as Abu Hureyra with thousands of people[10], and medium-small settlements of hundreds of individuals like Tell Halula[8]. One reason for the decline in population may have been a reduction in the carrying capacity of the basic agricultural system, namely cereal farming[11].

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