

ANTHROPOLOGY

Anthropogenic changes to the Holocene nitrogen cycle in Ireland

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Humans have always affected their ecosystems, but finding evidence for significant and lasting changes to pre-industrial landscapes is rare. We report on human-caused changes to the nitrogen cycle in Ireland in the Bronze Age, associated with intensification of agriculture and animal husbandry that resulted in long-term changes to the nitrogen isotope values of animals (wild and domesticates) during the Holocene. Major changes to inputs and cycling of soil nitrogen occurred through deforestation, land clearance and management, and more intensive animal husbandry and cereal crop cultivation in the later Bronze Age; after this time, the Irish landscape took on its current form. Within the debate concerning the onset of the Anthropocene, our data suggest that human activity in Ireland was significant enough in the Bronze Age to have long-term impact, thereby marking a profound shift in the relationship between humans and their environment.

INTRODUCTION

Humans are now recognized as having fundamentally altered the global nitrogen (N) cycle over the last 150 years. These changes have been primarily due to the transformation of unreactive molecular nitrogen to various reactive forms through the chemical synthesis of nitrogenous fertilizers and through fossil fuel combustion (1). The unprecedented industrial creation of reactive N through these processes has had widespread implications throughout the biosphere (1). Although comparatively little attention has been paid to the potential modification of biogeochemical cycles by ancient human populations, several studies have documented highly localized modifications of freshwater ecosystems by Arctic hunter-gatherers and northern horticulturalists (2, 3). Furthermore, it has been suggested that the Anthropocene era actually started in the Middle Holocene due to increased generation of CH₄ from ancient rice agriculture and animal husbandry in China (4), although this view has been disputed (5). Using N isotope compositions from Irish terrestrial mammals from the Late Pleistocene through to the Late Holocene, we demonstrate that human activity has had significant impact on the N cycle (at the soil-plant level) in Ireland as a result of widespread transformation of the landscape, which became dominated by human agropastoral activities.

Anthropogenic and natural phenomena influence the terrestrial N cycle and can, in turn, have a large impact on the $\delta^{15}\text{N}$ values of nutrients (in soils), producers (plants), and their consumers (animals) (6). Factors such as climate change, deforestation, and agricultural intensification can alter the biogeochemical processes responsible for large N isotope fractionations in the N cycle (7). Recent research points to three primary factors influencing the natural abundance of ^{15}N in soils and plants: mycorrhizal community composition, N cycle openness, and N inputs (quantity and isotopic composition) (7). These factors are nec-

essarily interrelated as increasing the input of labile N (through the application of fertilizers for example) will push the N cycle to be more prone to preferential ^{14}N loss through denitrification and ammonia volatilization (more “open”). Moreover, the mycorrhizal associations present are influenced by N mineralization and immobilization rates, which are integral components of determining the degree of N cycle openness (8). Human activity should primarily affect those factors controlling the N cycle through additions of higher levels of mineralized and highly labile N (that is, fertilization of fields and increased presence of livestock on pastures) and through management processes that serve to increase the rate of N cycling (for example, tilling of fields).

Here, we use animal bone collagen $\delta^{15}\text{N}$ values ($n = 757$) from a geographically wide range of sites ($n = 90+$) spanning the Irish Holocene (Fig. 1) to explore long-term patterns in N isotopic variation. These data are viewed as a record of broadscale anthropogenic impacts on Ireland’s terrestrial N cycle and soil nutrient dynamics. Results document a fundamental shift in animal $\delta^{15}\text{N}$ values across Ireland during the later Bronze Age [ca. 3000 calibrated years before present (cal BP)], coincident with a sharp rise in evidence for woodland clearance and cultivation from other paleoenvironmental records (9).

CONTEXT

Ireland lies at the northwestern fringe of Europe, and this unique geographical position has helped structure Irish natural and cultural landscapes. Ireland was ice-free until about 32,000 cal BP, but during the Last Glacial Maximum (26,000 to 19,000 cal BP), the island had likely become completely glaciated (10). With some low-lying areas fully deglaciated by 15,000 cal BP, it again became possible for plants and animals to return and recolonize (11). Lower sea levels during the last Ice Age had connected Ireland to Britain and Europe by a land or ice bridge, but with rising sea levels, at about 16,000 cal BP, Ireland again became an island, hindering its biological recolonization (12).

Very little is known about Ireland’s first human inhabitants before the end of the last Ice Age (ca. 11,500 to 12,000 cal BP); however, recent evidence suggests that Paleolithic human occupation had begun by at least 12,500 cal BP (13). More evidence for human activities is available from the Irish Mesolithic period (10,000 to 6000 cal BP) (14). The archaeology of Irish Mesolithic hunter-gatherers suggests low population densities with a focus on coastal, lacustrine, and riverine locations and

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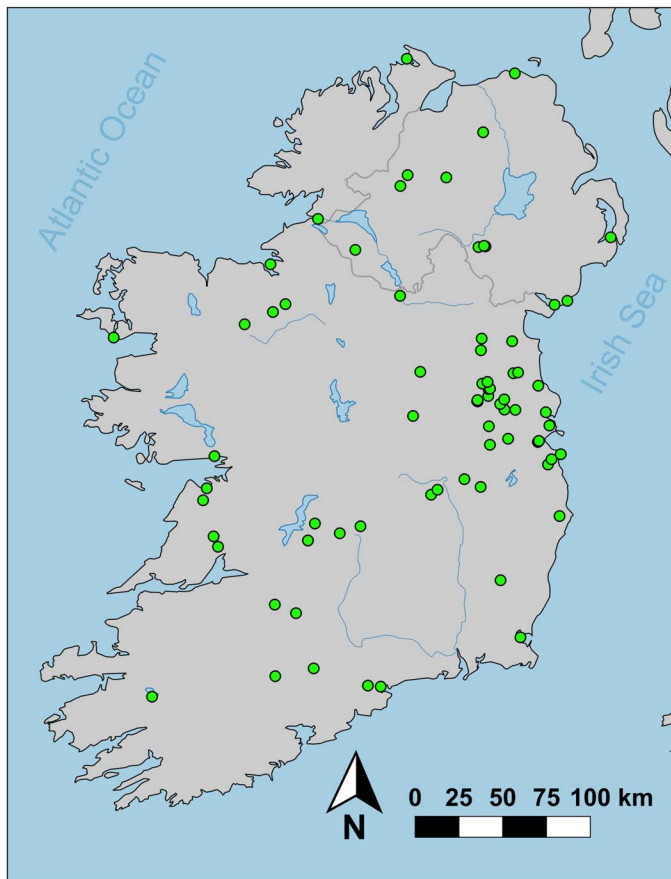


Fig. 1. Map showing locations (see table S4) from which samples were retrieved.

resources, but also drawing on a broad spectrum of local terrestrial plant and animal resources (14). Ireland hosted few, if any, large mammals at this time, and it is thought that Mesolithic people introduced wild boar (*Sus scrofa*) to the island (15). The Neolithic (6000 to 4500 cal BP) is marked by the introduction of domestic plants (cereal cultivars) and animals [cattle (*Bos taurus*), sheep (*Ovis aries*), goats (*Capra hircus*), and pigs (*Sus domestica*)] and additional wild species including red deer (*Cervus elaphus*), as well as new forms of shelter and monumental stone architecture, most notably a proliferation of chambered tombs (16). With crop- and animal-based agriculture, settlement increased significantly, but population levels may have tapered off toward the later Neolithic (17). The Irish Bronze Age (4500 to 2500 cal BP) is well known for its copper, bronze, and gold metal work (18). Ireland's Bronze Age archaeological record is characterized by an increasing number of sites, notably burials in the Early Bronze Age (4500 to 3500 cal BP), and later a proliferation of evidence for settlement (especially the mound-pit structures known as *fulachtaí fiadh*), track ways, and mortuary and ritual architecture, as well as intensified deforestation and agropastoral land use (9). Archaeological and paleoenvironmental evidence for the Iron Age (2500 to 1600 cal BP) is relatively sparse in comparison with earlier and later periods (19) but suggests fluctuating levels of agropastoral and land-clearing activities (20). The medieval period in Ireland (1600 to 350 cal BP) again saw significant population increases, along with the introduction of urbanization. The relative importance of livestock-keeping versus arable agriculture varied by time and geographical location. Livestock, particularly cattle, were dominant up to the 9th century AD, with a shift

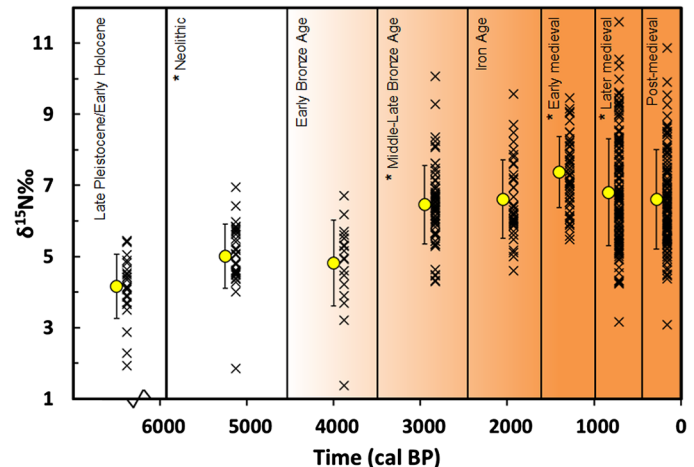


Fig. 2. Herbivore $\delta^{15}\text{N}$ values from table S2 plotted by chronological period. Asterisks show statistically significant difference between means of indicated time period and preceding time period.

toward more sheep and more arable agriculture later in the early medieval period. In the later medieval period (750 to 350 cal BP), an east-west divide was significant, with a pastoral economy more important in the wetter, poorer-quality lands of the west. The later medieval and post-medieval periods also saw significant deforestation and continued expansion of lands used for both arable and pastoral agriculture (21).

RESULTS

Stable nitrogen isotope ratios from bone collagen from archaeological wild ($n = 54$) and domestic ($n = 435$) herbivores and omnivores ($n = 178$) are presented in tables S1 and S2. Figure 2 shows a small but significant (Mann-Whitney U test, $U = 142.5$, $P < 0.001$) increase in average herbivore $\delta^{15}\text{N}$ values [$+0.8$ per mil (‰)] between the Late Pleistocene/Early Holocene ($n = 24$, $+4.2 \pm 0.9\text{‰}$) and the Neolithic ($n = 29$, $+5.0 \pm 0.9\text{‰}$), as has been observed elsewhere in Europe (22). Herbivore $\delta^{15}\text{N}$ values remain relatively low into the Early Bronze Age ($n = 19$, $+4.8 \pm 1.2\text{‰}$) but increase significantly (Student's t test, $t_{79} = -5.55$, $P < 0.001$), by nearly 2‰, during the transition to the Middle-Late Bronze Age ($n = 62$, $+6.5 \pm 1.1\text{‰}$). Average herbivore $\delta^{15}\text{N}$ values remain elevated during the Iron Age ($n = 40$, $+6.6 \pm 1.1\text{‰}$) and increase slightly during the early medieval ($n = 54$, $+7.3 \pm 1.0\text{‰}$), after which there is a small decline into the later medieval ($n = 134$, $+6.8 \pm 1.5\text{‰}$) and post-medieval ($n = 127$, $+6.6 \pm 1.4\text{‰}$). Although sample sizes for wild herbivores are relatively small for later time periods, a similar trend of increasing $\delta^{15}\text{N}$ values is observed through time (table S3 and fig. S2), suggesting that this baseline shift in herbivore bone collagen $\delta^{15}\text{N}$ reflects a broad change in the nitrogen isotopic compositions of plants and soil nutrients in Ireland's terrestrial ecosystem. There were also significant differences among the time periods examined for pigs [one-way analysis of variance (ANOVA), $F_{5,171} = 24.67$, $P < 0.001$; fig. S1]. The $\delta^{15}\text{N}$ values for pigs in the Neolithic and Early Bronze Age ($n = 15$, $\delta^{15}\text{N} = +5.4 \pm 0.6\text{‰}$) are significantly lower than those in the Late Bronze Age ($n = 35$, $\delta^{15}\text{N} = +6.5 \pm 0.9\text{‰}$, $P < 0.001$). The $\delta^{15}\text{N}$ values for pigs in all subsequent periods are higher than those during the Neolithic and Early Bronze Age, and this difference is statistically significant for the early medieval ($n = 20$, $\delta^{15}\text{N} = +8.4 \pm 0.9\text{‰}$, $P < 0.001$), later medieval ($n = 20$, $\delta^{15}\text{N} = +7.6 \pm 1.2\text{‰}$, $P < 0.001$), and post-medieval ($n = 28$, $\delta^{15}\text{N} = +8.2 \pm 1.5\text{‰}$, $P < 0.001$) periods.

DISCUSSION

As primary consumers, herbivore $\delta^{15}\text{N}$ values are directly linked to those of plant foods and therefore record changes in the terrestrial N cycle (6). Given the wide geographical distribution of this data set, we believe that the temporal shift observed in animal bone collagen $\delta^{15}\text{N}$ values during the Bronze Age is reflective of a process or processes occurring at a broad regional scale.

A large body of paleoenvironmental research on the Irish Holocene (23, 24) provides an excellent opportunity to evaluate which variables have driven N isotopic variation before and after the later Bronze Age shift observed in animal bone collagen $\delta^{15}\text{N}$ values (24). Pollen records from across the island document the introduction and intensification of agricultural and pastoral activities throughout the Irish Holocene from the Neolithic onward (25). Pollen records show variable but overall sustained low-level human landscape management after the Early Neolithic and continuing into the Early Bronze Age (16). A large number of pollen records then demonstrate a significant rise in human activity during the Mid-Late Bronze Age, as indicated by decreases in the frequency of arboreal pollen and increases of indicators for human disturbance such as *Plantago lanceolata*, Cerealia-type, and Poaceae pollen, as well as charcoal (9). This shift marks a significant reduction in woodland cover and increases in pastoral and arable farming activity, as well as possibly burning. These changes would contribute to general enrichment in soil ^{15}N through (i) a shift from ectomycorrhizae (primarily woodland associated) to arbuscular mycorrhizae (pasture and cultivar associated) as the dominant symbiotic partners for plants; (ii) promotion of N cycle openness through deforestation, soil tillage, and animal trampling; and (iii) increasing N inputs from agricultural by-products (for example, animal waste) (7). The fact that the isotopic shift is present in wild and domestic herbivore $\delta^{15}\text{N}$ values is significant because it suggests that changes in Ireland's terrestrial N cycle extended across a broader region, including both wooded and unwooded "wild" and agropastorally managed land. Human activities could influence wild fauna $\delta^{15}\text{N}$ values in two ways. First, the N isotopic composition of plants and wild fauna in unmanaged areas may have been influenced by nutrient inputs from adjacent land managed for agropastoral and arable uses. Second, because soil ^{15}N enrichments can persist for long periods (7), wild herbivores inhabiting fallow or abandoned areas that were previously heavily managed by humans should have higher $\delta^{15}\text{N}$ values.

Paleoclimatic data for the Irish Holocene show spatial and temporal variability (24), and it is possible that long-term warming could also have contributed to increased N cycle openness. Potential climatic influences on variation in animal $\delta^{15}\text{N}$ values appear to be less important, however, because there is no indication that prevailing precipitation and temperature conditions in Ireland have remained significantly warmer and drier from the later Bronze Age to the present. There is a significant body of well-dated evidence that has demonstrated that parts of Ireland experienced cooler, wetter conditions at the end of the Late Bronze Age, transitioning into the Iron Age (26). Later climatic fluctuations, including both drier and wetter periods, have occurred throughout the Iron Age and historical periods (27), but this climatic variation is not temporally consistent with the patterns observed in Iron Age, medieval, and post-medieval animal $\delta^{15}\text{N}$ values. Humans may also have altered Ireland's plant communities and woodland cover through reintroductions of species such as red deer (*C. elaphus*), which had disappeared from the island during the last Ice Age and were reintroduced in the Neolithic (28), and introductions of species such as fallow deer (*Dama dama*) during the medieval (29). These events also do not correspond temporally with changes observed in later Holocene Irish fauna $\delta^{15}\text{N}$

values and therefore would not have been a primary cause of landscape changes that influenced the N cycle at a regional scale. In this context, we believe that an increase in the breadth and intensity of human landscape use (from settlement, agriculture, and pastoral activity), rather than environmental factors, underlies the fundamental change to Ireland's N cycle that we have observed in Irish Holocene animal $\delta^{15}\text{N}$ values.

This study is the first analysis of isotopic variation in animals throughout the Holocene. Cumulatively, results show that anthropogenic woodland clearance, agropastoral activities, and land management have left a durable, long-term signature on the N isotope composition of soil, plants, and animals in Ireland. The implications of these findings, however, are global; the effect of human activity on soil N isotope composition may be traceable wherever humans have extensively modified landscapes for agriculture, albeit possibly much more subtly and gradually and over a longer span of time than has been observed here for Ireland. For this reason, our findings have significant potential to serve as a model for future research, using large archaeological faunal isotopic time series in other agricultural areas around the globe to reveal fundamental changes in the way that ancient societies have affected terrestrial N cycling.

In the context of debate about the onset of the Anthropocene, an important question is whether preindustrial human activities, particularly as detected through the archaeological record, should be included as defining factors (30). With respect to isotopic research, this topic has generated considerable discussion (31) about the importance of more recent changes to the global carbon (C) and N cycles. Significant efforts have been made, for instance, to understand the impact of large 20th-century N contributions, which are causing a global change in N isotope composition (32). Our results demonstrate that meaningful human alterations of Earth's biogeochemical cycles, at least at the regional scale for N cycling, are not unique to the industrial era. Our data show that humans have been significantly affecting the N cycle at a large scale for millennia. Anthropogenic alteration of the N cycle in prehistory would not necessarily have occurred synchronously at a global scale and so may not be considered as a potential marker in ongoing debates about defining the onset of the Anthropocene; however, it nonetheless represents a profound development in the way humans have interacted with their environments at the nutrient level.

Anthropogenic alterations of the N cycle, such as those observed during the Bronze Age in Ireland, represent an unprecedented shift in the scale and nature of ancient human impacts on the environment and therefore have significant implications for how we understand the way past agropastoral societies caused environmental change. The N cycle plays a vital role in nutrient dynamics at the soil-plant level, ultimately facilitating biogeochemical processes in the production, transfer, and recycling of key building blocks for plants and their consumers. Effective management of soil nutrient dynamics has been a prerequisite for sustained agricultural success throughout human history. In turn, the increased carrying capacity conferred by agricultural success has often been essential to human population growth and the development of complex, hierarchical societies. In that context, we can better understand the turning point at which past societies started to substantially restructure their environments, reorganizing them at a fundamental molecular level for the purpose of supporting growing human needs, by establishing where and when human activities began to change the terrestrial N cycle at a broad scale. Our data indicate that human impacts on the N cycle, as revealed through isotopic analyses of a long time series of archaeological animal remains, provide a convenient and direct

indicator for when agriculture and other human activities began to create such a detectable, long-term change to environmental nutrient dynamics. In particular, issues related to agricultural intensification and fertilization have also been explored using functional weed ecology (33) and stable isotopic compositions of macrobotanical remains (34) but have been spatially limited to the extent of the cultivated fields or archaeological sites being investigated. In contrast, isotopic analyses of animal bone collagen provide a long-term, time-averaged record of multiple years of dietary intake and therefore offer a broader spatio-temporal perspective.

There has been considerable debate about what types and intensities of human activity portend the fundamental economic, social, and technological shifts that have made humans a dominant driver of environmental processes at local, regional, and global scales. This discussion has not only guided debate on the relevance of archaeology to defining and understanding the Anthropocene but has also played a key role in structuring larger anthropological research questions on the spread of agriculture, the rise of social complexity, and the collapse of societies (35). Important archaeological indicators of these processes include evidence for the presence of cultivars and domesticates, soil erosion, and forest clearance, as well as warfare, monumental architecture, and social inequality. While these indicators provide clear evidence for fundamental changes in the nature of human societies at local and regional scales, from an ecological and nutritional perspective, they do not necessarily give an indication of the timing and intensity of human impacts on the environment or the extent to which humanity's activities drove broader ecosystem processes. In this context, evidence for the timing when humans began to significantly affect terrestrial nutrient dynamics at a local and regional scale, as we have shown in Bronze Age Ireland, can also provide a marker for the tipping point at which human societies changed paths to become drivers of ecosystem processes and, in turn, redefine their relationship with the environment.

MATERIALS AND METHODS

Sample description

Faunal samples ($n = 712$) from at least 90 (this number is the minimum estimate as several extinct Irish Giant Deer from natural history collections do not have associated provenance information) archaeological and natural history sites in 16 Irish counties were collected from private-sector archaeology companies, universities, and museums. Data for 45 additional animals were sourced from the literature (table S2) (28, 36–38). Species identifications were reconfirmed by an archaeozoologist (F.B.). General chronological parameters were established based on information from radiocarbon dates and archaeological context details from publications, unpublished site reports, and communications with excavators. Samples were selected with a view to maximizing geographical coverage in each temporal period. Where possible, an effort was made to avoid sampling younger individuals that may show ^{15}N enrichment due to milk feeding (39); however, relatively large sample sizes mitigate this issue.

Sample preparation

Bone samples were demineralized in 0.5 M HCl and rinsed to neutrality in type I ultrapure water. Where base-soluble contaminants (that is, humic acids) were apparent, samples were treated with 0.1 M NaOH in an ultrasonic bath repeatedly (solution refreshed every 15 min) until the solution remained clear. Treated samples were rinsed to neutrality again

in type I water and then solubilized in 10^{-3} M HCl (pH ~ 3) over 48 hours in an oven at 75°C . Collagen solutions were then filtered using 45- to 90- μm mesh filters (Elkay Laboratory Products) and 30,000 molecular weight cutoff filters (Pall Corporation), and the $>30\text{-kDa}$ fraction was then frozen and lyophilized. Sample integrity was evaluated using collagen yield, C/N ratio, and elemental percent criteria (40). Of 712 samples, 622 (88%) produced acceptable values for collagen quality indicators.

Stable isotope analysis

Isotopic analyses were performed in duplicate. Collagen samples (0.5 mg) were combusted in tin capsules in an Elementar vario MICRO cube elemental analyzer coupled to an Isoprime isotope ratio mass spectrometer in continuous flow mode at the University of British Columbia (UBC), Vancouver, Canada. Isotopic compositions were calibrated relative to ambient inhalable reservoir using glutamic acid standards USGS40 ($\delta^{15}\text{N} = -4.52\text{‰}$) and USGS41 ($\delta^{15}\text{N} = +47.57\text{‰}$) at UBC.

Statistical comparisons

To assess whether levels of $\delta^{15}\text{N}$ changed over time within herbivore (Fig. 2) and omnivore (fig. S1) data sets, normality was first assessed using a Shapiro-Wilk test. Within data sets where all time period groups were normally distributed (that is, omnivores), a one-way ANOVA was used. For the ANOVA, homogeneity of variance was assessed using Levene's test, and if the variances were unequal among groups, then a post hoc Dunnett's T3 test was performed; otherwise, a Tukey's post hoc test was performed. Within data sets that contained one or more non-normally distributed time period groups (that is, herbivores), a t test was used to compare time period groups that were both normally distributed, and a Mann-Whitney U test was used for all other comparisons that included a time period group that was not normally distributed.

SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at <http://advances.sciencemag.org/cgi/content/full/4/6/eaas9383/DC1>

fig. S1. Omnivore (pig) $\delta^{15}\text{N}$ values from table S2 plotted by chronological period.

fig. S2. Wild (blue "x") versus domestic (black "x") herbivore $\delta^{15}\text{N}$ values from table S2 plotted by chronological period.

table S1. Summary of contextual information and average $\delta^{15}\text{N}$ values ($\pm 1\sigma$) for herbivores and omnivores by time period.

table S2. Contextual information and stable isotope and elemental concentration data for samples analyzed in this study and those sourced from literature.

table S3. Average $\delta^{15}\text{N}$ values ($\pm 1\sigma$) from table S2 for wild and domestic herbivores by time period.

table S4. Latitude and longitude for sites in this study.

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