

Deforestation caused abrupt shift in Great Lakes nitrogen cycle

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Abstract

Despite the longstanding significance of North America's Great Lakes, little is known about their preindustrial ecology. Here, we report on when and how humans first became a main driver of Lake Ontario's nutrient dynamics. Nitrogen isotope analyses of archaeological fish show that, prior to the 1830s, Lake Ontario's nitrogen cycle and the trophic ecology of its top predators had remained stable for at least 800 yrs, despite Indigenous and historical European agricultural land management across the region. An abrupt shift in the nitrogen isotope composition of Lake Ontario's fish community is evident in the early to mid-19th century and reflects the initiation of industrial-scale forest clearance. These data show how the nitrogenous nutrient regimes of even the world's largest freshwater ecosystems can be highly sensitive to short-term watershed forest cover disturbances and indicate a profound shift in the relationship between humans and their environment.

Throughout the 20th century, increased anthropogenic contributions of reactive nitrogen to freshwater environments have profoundly altered aquatic biogeochemical cycles and broader nutrient dynamics, resulting in widespread ecosystem dysfunction, loss of biodiversity, and degraded water quality (Smith and Schindler 2009). While numerous studies have documented how increasing anthropogenic nitrogen inputs are incorporated into aquatic food webs, promoting cultural eutrophication and altering trophic dynamics in the recent past (Schindler et al. 2006), less consideration has been given to how historical and preindustrial populations have impacted aquatic ecosystems.

In the context of recent debate about the timing of the origins of the Anthropocene (Lewis and Maslin 2015), in which past societies are considered as potential architects of the first broad-scale environmental changes, the role of humans as drivers of biogeochemical processes such as the nitrogen cycle is becoming increasingly important (Hadley et al. 2010; Kintigh et al. 2014; Guiry et al. 2018).

For conservation and restoration efforts in freshwater ecosystems where cultural eutrophication caused by increased nutrient loading poses one of the most significant threats globally (Smith and Schindler 2009), long-term retrospectives that document when and how human activities first began to alter natural ecosystem processes, and the nitrogen cycle in particular, could provide a valuable framework for evaluating which modern human activities pose the greatest risks (Humphries and Winemiller 2009; Canfield et al. 2010). Paleolimnological proxy indicators based on the physical, biological, chemical, and isotopic compositions of sediments have long been used to establish trends in past aquatic productivity and water quality (Beeton 1965; Schelske et al. 1983; Hodell and Schelske 1998; Jeffers et al. 2015), but, because of

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potential taphonomic issues (Anderson 2014; Lu et al. 2014) and because these approaches usually do not incorporate consumers, they are unable to measure the direct impacts of anthropogenic nitrogen loading on nutrient dynamics in the wider biotic community (e.g., invertebrates, fish, birds; although for a growing literature on invertebrate analyses, see Perga 2010; Perga et al. 2010; van Hardenbroek et al. 2010; Frossard et al. 2014; Schilder et al. 2017; Anas et al. 2019). Moreover, while many studies using isotopic analyses of museum-archived vertebrate tissues have been able to make important contributions to understanding how biotic communities have responded to human-caused environmental change during the 20th century (Fera et al. 2017), a lack of suitable specimens from early historical and preindustrial time periods has, in most cases, prevented analyses of longer-term environmental variation in vertebrate taxa (Szpak et al. 2018). In this context, stable nitrogen isotope and other analyses of ancient fish remains from archaeological repositories can provide an invaluable opportunity to gain direct insight into how food web and nutrient dynamics within preindustrial ecosystems functioned and responded to impacts from human activities at varying temporal and spatial scales (Guiry et al. 2016b).

Using isotopic compositions of fish bone or scales has some interpretive advantages relative to more commonly analyzed materials, such as bulk sedimentary organic matter, because fish stable nitrogen isotope composition is linked firmly with a known ecological point (i.e., a particular species with well-understood ecology and trophic affinity) and spatiotemporal framework (Trueman and Moore 2007) and can be easily parsed to remove diagenetically altered samples using robust quality-control criteria (DeNiro 1985; Szpak 2011). This is important because, in contrast to analyses of bulk sedimentary organic matter, which is composed of a spatially, temporally, and biologically heterogeneous mixture of sources of detritus (Lu et al.

2014), with fish bone or scales, the ability to compare “taxonomically anchored” isotopic patterns across different parts of a food web will enable much higher-resolution interpretations of environmental change at both the level of nutrient dynamics and the level of broader food web structures. Moreover, with respect to measuring the isotopic compositions of ancient organic materials, the ability to purify collagen extracted from bone (through pretreatment steps outlined below) and assess the extent to which its constituent carbon and nitrogen are biogenic (e.g., using well-established and precise C : N_{Atomic} criteria; DeNiro 1985; Szpak 2011) provides an additional advantage relative to other important paleontological materials (e.g., chitinous remains from subfossil invertebrates; Anas et al. 2019) and makes bone and scale collagen particularly well suited for intersite comparisons of isotopic variation in biota across time and space. Furthermore, because collagen from bones and scales is constructed and remodeled slowly throughout the life of an organism, the isotopic composition of collagen can provide a lifetime average record of dietary intake and environmental conditions that is less susceptible to seasonal or short-term idiosyncratic shifts in behavior (Hobson and Clark 1992). Bone collagen is therefore well suited for reconstructing long-term trends in biology and ecology (Bump et al. 2007) and can provide an additional perspective with potential to complement and build on the success of previous, well-established paleolimnological proxy indicators (e.g., physical, chemical, and isotopic compositions of sediments and invertebrates; Beeton 1965; Schelske et al. 1983; Hodell and Schelske 1998; Perga 2010; Jeffers et al. 2015).

We use stable nitrogen isotope compositions and peptide mass fingerprinting (Zooarchaeology by Mass Spectrometry, hereafter, ZooMS) of late Holocene (900–2015 CE) fish bone collagen from a large number of sites associated with Lake Ontario (Fig. 1), the most easterly of the Laurentian Great Lakes (hereafter, Great Lakes), to document long-term trends in regional biogeochemical cycles. Results show that, in comparison with the industrial era, the nitrogen cycle and trophic structure of this Great Lakes ecosystem remained remarkably stable until the 1830s, despite millennia of Indigenous agricultural and other land management, decades of European settlement, and climatic fluctuations. After this time, increased N-loading from forestry- and agriculture-induced soil erosion caused an unprecedented and abrupt bottom-up shift throughout the entire aquatic ecosystem of Lake Ontario.

Context

With more than 20% of the world’s surface freshwater, North America’s Great Lakes are of tremendous ecological, economic, and social interest (Sterner et al. 2017). Since monitoring programs began, the Great Lakes have experienced significant ecological and chemical changes in response to pollution, hydrological controls, and species invasion

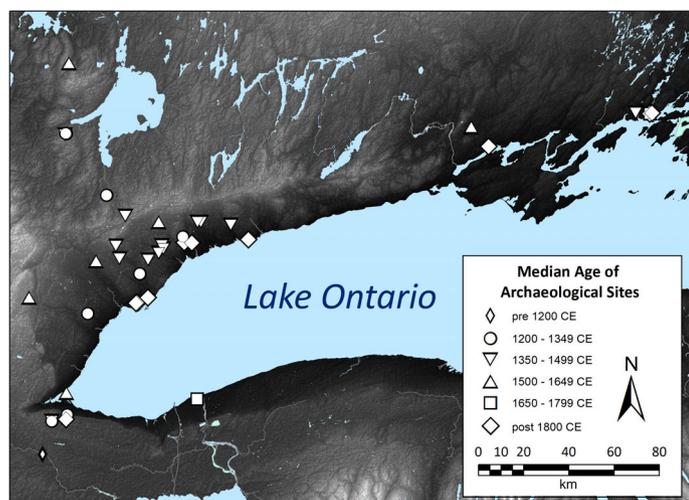


Fig. 1. Map of study area showing locations of archaeological sites from which fish bone samples were collected. Map data from NASA and ESRI.

(Allan et al. 2013; Stewart et al. 2016). Paleolimnological studies show that recent industrial activities have had profound impacts on Great Lakes productivity (Beeton 1965; Schelske et al. 1983, 1988; Schelske and Hodell 1991, 1995; Hodell and Schelske 1998; Meyers 2003; Jeffers et al. 2015), especially through increased phosphorus and nitrogen loading that led to eutrophication of Lake Ontario and Lake Erie by the 20th century (Hodell and Schelske 1998; Lu et al. 2010). With respect to nutrient dynamics, particularly the nitrogen cycle, which plays a vital role in primary production in naturally oligotrophic ecosystems such as Lake Ontario (Lean and Knowles 1987; Leggett et al. 2000), the extent to which these changes represent a departure from longer-term trends in broader food web structure and nutrient dynamics remains unclear (Sterner et al. 2017; Supporting Information 1). But this extent could be determined through isotopic reconstructions of past nutrient and food web dynamics using historical or archaeological fish specimens, as these can provide a complementary isotopic record for ancient environmental variation.

Human activities have modified the cycling of nitrogen in aquatic environments across the globe, contributing to the ongoing threat of cultural eutrophication (Gruber and Galloway 2008; Smith and Schindler 2009). The majority of these impacts occur when anthropogenic nitrogen inputs and/or chemical or physical changes to an environment alter the conditions regulating the nitrogen cycle, thereby disrupting the flow of nitrogenous nutrients through an ecosystem (Kendall et al. 2007). Modeling how nitrogen moves into and through aquatic ecosystems is, therefore, of considerable importance for conservation efforts seeking to restore heavily impacted watersheds (Denk et al. 2017). With respect to broadscale human impacts, such factors as climate change, acid rain, nutrient loading, and invasive species introductions can significantly alter the biogeochemical processes that balance an aquatic ecosystem's nitrogen cycle and often, in turn, cause detectable changes in the isotopic composition of nitrogen pools that are available to support aquatic life (Lake et al. 2001; Anderson and Cabana 2005; Botrel et al. 2014). Stable nitrogen isotopic compositions of consumer tissues show a stepwise increase, by roughly 3–4‰, in $\delta^{15}\text{N}$ values with each trophic level step up in a food web and have therefore traditionally provided a powerful indicator for food web interactions in ecology and archaeology (Post 2002). However, because any impact on the $\delta^{15}\text{N}$ of aquatic nitrogen pools at the base of a food web (i.e., in various pools of dissolved inorganic nitrogen [DIN]) is passed up the trophic ladder to producers (phytoplankton) and their consumers (invertebrates and fish), isotopic analyses of tissues from aquatic producers and consumers can also serve as a highly integrative indicator for anthropogenic impacts on nutrient dynamics, in addition to food web structure, in aquatic environments (Hoffman et al. 2012; Morrissey et al. 2013). While a majority of isotopic research has approached the question of changing freshwater nutrient dynamics through analyses of sedimentary organic

matter (e.g., Talbot 2001; Dubois et al. 2018), a growing number of studies are demonstrating that $\delta^{15}\text{N}$ compositions of invertebrates and fish can provide a highly sensitive record for environmental change (e.g., Perga et al. 2010; Lumb and Johnson 2012; Fera et al. 2017; Anas et al. 2019).

The natural abundance of ^{15}N in DIN is largely controlled by three factors (for reviews, see Kendall 1998; Talbot 2001; Finlay and Kendall 2007; Kendall et al. 2007; Guiry 2019): productivity (Schelske and Hodell 1991; Hodell and Schelske 1998), environmental conditions (temperature, oxygenation, and pH; Knowles 1982; Finlay and Kendall 2007), and nitrogen inputs (quantity and isotopic composition; Heaton 1986; Lake et al. 2001). These factors are necessarily interrelated because, for instance, increasing the input of reactive nitrogen can increase productivity, and this can affect environmental conditions that regulate other parts of the nitrogen cycle (Finlay and Kendall 2007). Moreover, nitrogen-limitation rates can influence plankton community composition, which is an integral component of determining the presence of nitrogen-fixing bacteria (Gu 2009) that can further modify the isotopic composition of an aquatic ecosystem by introducing isotopically distinctive atmospheric nitrogen. Human activity should affect those factors controlling the nitrogen cycle and its isotopic composition (for review, see Guiry 2019) by: (1) contributing reactive nitrogen (i.e., through inputs directly from sewage, agriculture, and soil erosion—typically, but not always, leading to elevated biota $\delta^{15}\text{N}$; Anderson and Cabana 2005; Morrissey et al. 2013), which itself leads to increasing productivity (creating greater nitrogen demand and thereby possibly promoting the importance of N-fixing algae—leading to higher and lower biota $\delta^{15}\text{N}$, respectively; Pennock et al. 1996; Gu 2009), and (2) changing chemical conditions, such as pH and oxygen levels, that regulate transformations between important forms of reactive nitrogen (altering the dynamics for nitrification and denitrification—with potential to push biota $\delta^{15}\text{N}$ higher or lower; Collister and Hayes 1991; Sebilo et al. 2006).

With the longest history of European and Indigenous agricultural land management and settlement among the Great Lakes, Lake Ontario and its watershed are an ideal context in which to investigate how large-scale human activities can impact major freshwater environments over time (Hodell and Schelske 1998). Because Lake Ontario receives a large amount of its water from land runoff, it is particularly sensitive to broader ecological processes and human disturbances occurring across the watershed, particularly in terrestrial and riparian zones. Moreover, the significant and highly visible environmental degradation of Lake Ontario's watershed throughout the 20th century, as well as its close proximity to major population centers, has spurred considerable research into its ecological structure (e.g., Bogue 2001; Meyers 2003; Estep and Reavie 2015; Stewart et al. 2016). For this reason, the lake has a relatively well-documented ecological and biogeochemical history, with numerous isotopic studies of

modern fish identifying recent trends in community structure, trophic dynamics, and energy flows (Kiriluk et al. 1995; Lumb and Johnson 2012; Rush et al. 2012; Yuille et al. 2015; Colborne et al. 2016; Fera et al. 2017). The longest-term of these studies, however, only offers a 70-yr retrospective, one that postdates intensification of European settlement of Lake Ontario's watershed by more than 150 yr.

While a number of important isotopic studies have helped to document longer-term variability in Lake Ontario's nitrogenous nutrient dynamics (Supporting Information 1), these have focused on isotopic compositions of organic matter in lake sediments rather than specific taxa. With respect to the last millennium, the timeframe over which human land management would have intensified, most (95%) of these analyses focus on the period after the mid-19th century and show substantial change in the nitrogen isotopic composition of organic matter settling out of Lake Ontario's water column (Supporting Information 1 Fig. S1, Fig. 2; Hodell and Schelske 1998; McFadden et al. 2004). Only a small fraction (5%; $n = 8$ of 178) of analyses cover the early historical period (1600s–1850 CE), when European activities would have started impacting the region, and precontact period, when Indigenous farming and other land management activities were accelerating across the watershed (i.e., starting with the Late Woodland, ca. 900 CE). Moreover, only one lake sediment isotopic data set spans both time frames and is not consistent with the others (Supporting Information 1, Fig. 2). While these data provide an invaluable macro-scale perspective on general patterns in nutrient dynamics in Lake Ontario, they cannot be used to assess the consequences of shifting

nitrogenous nutrient regimes for the broader food web (invertebrates and fish). Moreover, the coarse temporal resolution of the lake sediment isotopic record prior to 1850 prevents detailed analyses of when the human activities began to impact the nitrogen cycle and to what extent early historical and Indigenous activities may have been important drivers for change. In this context, isotopic analyses of archaeological fish can provide a complementary line of evidence for nutrient dynamics and food web structure.

Materials and methods

Sample description

Sampling focused on three taxa in the family Salmonidae: lake trout (*Salvelinus namaycush*; $n = 222$), Atlantic salmon (*Salmo salar*; $n = 158$), and whitefishes (*Coregonus* spp.; primarily *C. clupeaformis* and *C. artedii*, see below; $n = 277$; Supporting Information 2 Table S1). These taxa were selected to provide a highly integrated record of changes to Lake Ontario's nitrogen cycle and trophic structure. The upper trophic position and longer lifespan (5–15 yr) as well as pelagic foraging and highly mobile behavior of these species means that their diets will incorporate a broad range of resources from across the lake (Rush et al. 2012; Guiry et al. 2016b; Fera et al. 2017), providing a time-averaged perspective on processes affecting the nitrogen cycle at a lake-wide, multiseasonal scale. A comparison of isotopic variation among three taxa, each with a different behavioral strategy and trophic position (Holm et al. 2009; Fera et al. 2017; Mumby et al. 2018), should make isotopic trends associated with dietary shifts at the species level

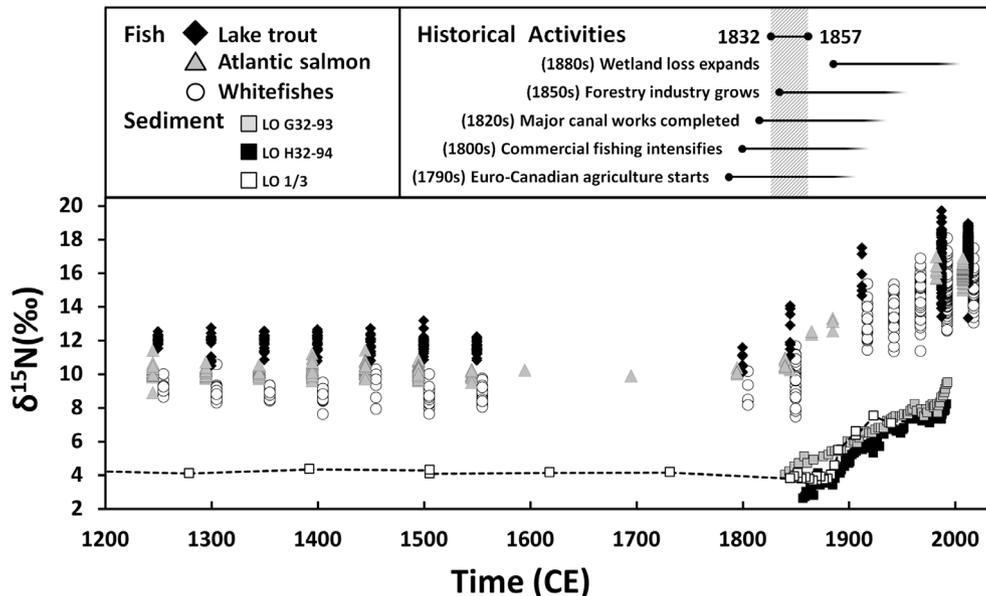


Fig. 2. Stable nitrogen isotope values for fish ($n = 1469$) vs. time. Data are binned by 50-yr (archaeological) and 25-yr (historical/modern) intervals. For fish data sourced from the literature (Kiriluk et al. 1995; van der Merwe et al. 2003; Lumb and Johnson 2012; Rush et al. 2012; Yuille et al. 2015; Colborne et al. 2016; Guiry et al. 2016b; Fera et al. 2017; Mumby et al. 2018). For sediment data sourced from the literature: LO G32-93, LO H32-94 (Hodell and Schelske 1998), and LO 1/3 (McFadden et al. 2004).

(as opposed to changes at the base of the food web) clearly discernible and, therefore, these data also give a long-term perspective on the stability of Lake Ontario's food web structure.

With the exception of nine Atlantic salmon samples, all archaeological specimens are from sites within the Lake Ontario watershed. As Lake Ontario is the only known source of Atlantic salmon, specimens from this species at sites outside the watershed were also included because they represent individuals originating from Lake Ontario. Where possible, archaeological fish bone specimens were selected based on minimum number of individual counts per archaeological context in order to avoid sampling the same individual multiple times. For all species, archaeological sampling efforts targeted bone specimens from adult-sized fish in order to exclude juveniles, who may feed at a lower trophic level. All specimens were examined by one or more of three zooarchaeologists (S.N.-H., A.L.H., T.J.O.) with specific expertise in the identification through comparative osteology of archaeological fish bones from relevant taxa in the Great Lakes region (Hawkins et al. 2019). The archaeological samples are largely derived from fish vertebrae, because they are more ubiquitous than fish cranial and pectoral girdle bones in the archaeological collections. Zooarchaeologists determined species for each specimen where possible, based on clear morphological differences between taxa. Due to overlaps in osteology and the possibility of hybrids, morphological examination of *Coregonus* vertebrae and many *Coregonus* cranial and pectoral girdle bones cannot be confidently used to assign species-level identification in the Lake Ontario watershed. However, of the five *Coregonus* species that are native to Lake Ontario, only two, lake whitefish (*C. clupeaformis*) and cisco (*C. artedii*), inhabit waters shallow enough to have been broadly accessible using fishing technologies (birch bark canoes and gill nets) available in the precontact past, and it is therefore highly likely that archaeological *Coregonus* of that time are composed of these taxa. Some bones in the genera *Salvelinus* and *Salmo* are also morphologically similar to each other, resulting in additional higher-level taxonomic identifications (including to *Salmo salar*/*Salvelinus namaycush* and Salmonidae). In cases where a species-level taxonomic identification was not achieved with a high degree of certainty, we undertook ZooMS analyses ($n = 292$) to confirm identifications, comparing the archaeological samples with reference sequences we generated for *S. salar*, *S. namaycush*, brook trout (*Salvelinus fontinalis*), round whitefish (*Prosopium cylindraceum*), *C. clupeaformis*, and *C. artedii* (Supporting Information 4 Table S1).

Dating for precontact specimens is based on published (Supporting Information 2 Table S2) radiocarbon dates, ceramic seriations, and reconstructed village occupation sequences (e.g., Williamson 2014), to occupations within 100-yr bins. Excellent chronological control for data from Indigenous archaeological sites reflects the highly standardized lifecycle in which settlements were built, occupied, decommissioned, and left behind in favor of relocation every

10–30 yr (Warrick 2008). The relative dearth of data from the seventeenth and eighteenth centuries reflects changes in Indigenous settlement patterns during this period. Reduced sample sizes in the 19th century reflect lower frequencies for fish bone at Euro-Canadian archaeological sites. Historical fish scale samples of adult specimens of known date were taken from museum-archived collections with assistance from an Ontario fish identification expert (E.H.). Analyses of scales from museum-archived *Coregonus* specimens include both *C. clupeaformis* and *C. artedii* because: (1) these two species likely account for the majority of archaeological specimens and will therefore be directly comparable; and (2) based on $\delta^{15}\text{N}$ values, they appear to have fed at the same trophic level in early 20th-century Lake Ontario. For data sourced from the literature ($n = 834$; Kiriluk et al. 1995; van der Merwe et al. 2003; Rush et al. 2012; Yuille et al. 2015; Colborne et al. 2016; Guiry et al. 2016b; Fera et al. 2017), fork length, when available, was used to exclude juveniles, which may feed at a lower trophic level.

Sample preparation

Samples weighing between 30 and 300 mg were cut from bone specimens, residual bone lipids were removed with a series of 2:1 chloroform: methanol ultrasonic baths (solution refreshed every 10 min until visible signs of reaction ceased), and the samples were then left to air dry (Guiry et al. 2016a). Bone samples were then demineralized in 0.5 mol L⁻¹ hydrochloric acid (HCl) for several days at 4°C and then rinsed to neutrality in Milli-Q water. Humic acids and other base soluble contaminants from the burial environment were removed from demineralized bone samples with successive treatments in 0.1 mol L⁻¹ sodium hydroxide in an ultrasonic bath (solution refreshed every 15 min until visible signs of reactions ceased) and then rinsed to neutrality in Milli-Q water (Szpak et al. 2017a). Bone samples were then solubilized in 10⁻³ mol L⁻¹ HCl in an oven at 65°C for 48 h. The resulting solutions were centrifuged, after which the solubilized collagen fraction was transferred to a new tube and then frozen and lyophilized.

The protein fraction of fish scale is also composed primarily of collagen, which is compositionally and isotopically comparable to fish bone collagen (Trueman and Moore 2007; Guiry et al. 2016b). Fish scales samples underwent a succession of three rinses in Milli-Q water in an ultrasonic bath for 5 min each (Guiry et al. 2016b). Scale samples were then soaked for 5 min in 1.0 mol L⁻¹ HCl in an ultrasonic bath to remove the mineral phase of the scale's external plate as well as any microscopic debris that may have become adhered to the external surfaces of the fish specimens during preservation and storage. Scale samples were then rinsed to neutrality in Milli-Q water in an ultrasonic bath and left to air dry. The historical fish specimens had been preserved using formalin fixation and then stored in ethanol. While this form of preservation is known to introduce small quantities of carbon,

which can slightly alter the stable carbon isotope composition of biological tissues, formalin and ethanol provide no new sources of nitrogen and therefore have a minimal influence the nitrogen isotopic composition of preserved fish scales. To confirm this, we also compared the percent carbon-to-nitrogen ratio (C : N) of historical scales with that of modern fish scales (see below; Szpak 2011) to establish that diagenetic nitrogen is not present.

Isotopic analyses

The stable nitrogen isotope composition as well as percent carbon and nitrogen was measured on 0.5 mg samples of bone and scale collagen using an Elementar Vario MICRO cube elemental analyzer coupled via continuous flow to an Isoprime isotope ratio mass spectrometer in the Archaeology Chemistry Laboratory at the University of British Columbia, Canada. Duplicate or triplicate analyses were performed on 14% of samples. For a full account of procedures used for calibration of isotopic values as well as isotopic uncertainty calculations, see Supporting Information 3. Analytical uncertainty for $\delta^{15}\text{N}$ measurements was +0.20‰ (Szpak et al. 2017b; Supporting Information 3). The following data quality criteria were applied for establishing the integrity of $\delta^{15}\text{N}$ measurements from bone and scale collagen (DeNiro 1985): (1) % carbon and % nitrogen values above 13% and 4.8%, respectively; and (2) atomic C : N values falling between 2.9 and 3.6.

ZooMS analyses

Collagen was rehydrated with approximately 100 μL 50 mmol L^{-1} ammonium bicarbonate per milligram and each digested with 0.4 μg sequencing grade trypsin (Promega, UK) overnight at 37°C. Initial attempts were made at directly spotting these digests through dilution in 0.1% trifluoroacetic acid (TFA; Buckley et al. 2018), but many required ZipTip purification with C18 solid phase extraction and rehydration in 0.1% TFA (Buckley et al. 2009). Samples were spotted onto a stainless-steel Matrix Assisted Laser Desorption Ionization (MALDI) target plate with equal volume of 10 mg mL^{-1} alpha-cyano hydroxycinnamic acid in 50% acetonitrile/0.1% TFA and allowed to air dry. Dried spots were then analyzed using a Bruker Ultraflex II MALDI mass spectrometer over the m/z range 700–3700 with up to 2000 laser acquisitions for each sample. Peptide mass fingerprints were then compared with those of standard reference material via the identification of genus- or species-specific biomarkers (Supporting Information 4).

Statistical analyses

For statistical comparisons of fish $\delta^{15}\text{N}$ values, each taxon (i.e., lake trout, Atlantic salmon, whitefishes) was compared separately by time periods binned by 100 (pre-1925 CE) and 25 (post-1925 to present) year groups (Supporting Information 5 Table S1) using PAST version 3.22. Bin timeframes were selected with a view to maximizing sample size per bin. To assess whether fish $\delta^{15}\text{N}$ values changed over time, we

first compared pre-1800 bins to one another to establish whether significant differences occur within each fish taxon between sequential time frames. After finding that no significant differences occur (Supporting Information 5 Table S2), we then grouped all pre-1775 (preindustrial) bins and compared this individually to all succeeding time periods (Supporting Information 5 Table S3). For each bin, normality of distribution was first tested using a Shapiro–Wilk’s test (Supporting Information 5 Table S1). For data sets where all time periods were distributed normally, a one-way ANOVA was used, and homogeneity of variance was assessed using a Levene’s test. A post hoc Dunnett’s T3 test or a post hoc Tukey’s HSD test was used to compare groups with variances that were equal or unequal, respectively. For data sets where not all time periods were distributed normally, Mann–Whitney U tests with Bonferroni corrected p values were used to compare groups.

Results

Of the samples with sufficient collagen ($n = 607$ of 662) for isotopic analyses, 96% (total $n = 582$) produced acceptable C : N, %C, and %N values satisfying quality control criteria. Interpretations also include previously published data from archaeological ($n = 68$; van der Merwe et al. 2003; Guiry et al. 2016b) and modern (post-1958 CE; $n = 819$; Kiriluk et al. 1995; Lumb and Johnson 2012; Rush et al. 2012; Yuille et al. 2015; Colborne et al. 2016; Fera et al. 2017; Mumby et al. 2018) specimens. ZooMS analyses of 290 samples confirmed or helped to refine 244 nonspecies taxonomic identifications provided through bone morphology analyses by zooarchaeologists (it confirmed the identification for 225 of these 244 specimens and refined it to the level of species for the remaining 19). In a further 20 cases, specimens were reassigned to another taxon, namely, 12 to lake trout (*Salvelinus namaycush*) (previously assigned with varying degrees of confidence to the species or genus level in *Salmo* [$n = 8$], *Salvelinus* [$n = 2$], or *Coregonus* [$n = 2$]); four to Atlantic salmon (*Salmo salar*) (previously identified with varying degrees of confidence as *Salvelinus namaycush*); four to whitefish (*C. clupeaformis* or *C. artedii*) (previously assigned with varying degrees of confidence to the species or genus level in *Salmo salar* [$n = 1$], *Salvelinus fontinalis* [$n = 2$], or *Salvelinus namaycush* [$n = 1$]). In 15 other cases, ZooMS could not reassign specimens (previously assigned with varying degrees of confidence to the Salmonidae family level [$n = 3$] or to the species or genus level in *Salmo* [$n = 4$], *Salvelinus* [$n = 1$], or *Coregonus* [$n = 4$]) to one of the four species and one genus used in this study. Based on variation in peptide mass fingerprints, these samples likely derive from at least eleven other species.

Stable nitrogen isotope compositions of bone and scale collagen from archaeological and historical lake trout ($n = 196$), Atlantic salmon ($n = 147$), and whitefishes ($n = 239$) are summarized in the Supporting Information 5 Table S1 and

presented in full in Fig. 2 and Supporting Information 2. Given the large and well-dated sample ($n = 1469$) from archaeological sites ($n = 42$) and from museum-archived specimens and contemporary fisheries research ($n \leq 50$ locations) across a broad geographical range, we believe that the temporal trends observed in fish bone and scale collagen $\delta^{15}\text{N}$ are representative of processes occurring in Lake Ontario's open water ecosystem at a regional scale.

Discussion and conclusion

1200–1830 CE

Fish $\delta^{15}\text{N}$ shows remarkably little variation between 1200 CE and 1830 CE (Fig. 2), despite centuries of Indigenous and European land management in the Lake Ontario watershed, and provides a unique perspective on the long-term stability of a Great Lakes ecosystem. Prior to the 19th century, mean $\delta^{15}\text{N}$ values binned by taxon and at 100-yr intervals vary by less than 0.4‰ and show no statistically significant differences (Supporting Information 5 Tables S1, S2). This long-term stasis, over at least 600 yr, suggests that the nitrogen cycle and nitrogenous nutrient inputs for Lake Ontario were highly stable during the past millennium. Analyses of two additional Atlantic salmon specimens (not shown in Fig. 2) from earlier archaeological deposits suggests that this stability extends further back in time to at least 900 CE. However, due to a paucity of samples from the earlier time period, discussion will focus on the post-1200 CE time frame. A comparison of mean $\delta^{15}\text{N}$ for pre-1800 bins ($n = 4$, see Supporting Information 5 Table S1) shows steady offsets between species: lake trout $1.6 \pm 0.2\text{‰} >$ Atlantic salmon $1.3 \pm 0.2\text{‰} >$ whitefishes. Replication of this pattern across all three taxa, encompassing multiple trophic levels, suggests that stability was also characteristic of interspecific trophic relationships throughout the broader pelagic food web during this period.

A wealth of historical and paleoenvironmental research on the Lake Ontario watershed (Beeton 1965; Schelske et al. 1983, 1988; Schelske and Hodell 1991, 1995; Hodell and Schelske 1998; Wood 2000; Bogue 2001; Meyers 2003; Estep and Reavie 2015; Jeffers et al. 2015) provides an excellent opportunity to contextualize and consider the implications of this long-term stability. Proxies for past environmental conditions in other, smaller lakes in the same region document anthropogenic disturbances, at least as early as the 13th century CE, resulting from Indigenous land clearance and farming activities (e.g., Ekdahl et al. 2004). These studies suggest that Indigenous land management practices, including the use of fire for forest clearance followed by crop cultivation (Munoz and Gajewski 2010), had a significant impact on soil erosion, sedimentation, and nutrient loading, causing early cultural eutrophication of aquatic environments in the region. European settlement of the Lake Ontario watershed began in the 17th century (Gentilcore 1984). Settlement expansion in

the 19th century was accompanied by intensive commercial fishing (1800 onward; Bogue 2001), diversion of the Niagara River and work on the Erie and Welland canals (1820s; McIlwraith 1976; Wood 2000), broad-scale land clearance for lumber and agriculture (1840s–1850s; Lower et al. 1938; Head 1975), and wetland loss due to mining and other activities (1880s; Bogue 2001). Paleolimnological studies suggest that productivity of Lake Ontario's phytoplankton community began to grow slowly with the first European settlement, with more substantial impacts occurring in the mid-19th century, probably in response to nutrient loading from soil erosion following deforestation (e.g., Schelske 1991; Hodell et al. 1998; Estep and Reavie 2015; Supporting Information 1). Despite clear historical and paleolimnological evidence for earlier anthropogenic impacts on nutrient dynamics (e.g., nitrogen and phosphorus loading, pollen species change) in other areas of the watershed (Duthie and Sreenivasa 1971; Schelske et al. 1983; Yang et al. 1993; Bunting et al. 1998; Ekdahl et al. 2004, 2007; Munoz and Gajewski 2010), archaeological fish stable nitrogen isotopic compositions demonstrate that Lake Ontario's nitrogen cycle and food web structure remained unchanged until the 1830s. In that context, and given the sensitivity of aquatic nitrogen cycles to human disturbances (Lake et al. 2001; Anderson and Cabana 2005; Botrel et al. 2014), these data underscore Lake Ontario's stability throughout much of the last millennium and demonstrate a degree of resilience at the level of the nitrogen cycle in large lake ecosystems to a variety of human impacts, even when sustained over long time spans.

The resilience of Lake Ontario's nitrogen cycle has important implications for understanding human impacts on freshwater ecosystems in the recent and more ancient past. It is now well known that industrial activities over the past 150 yr have fundamentally altered global nutrient distributions and biogeochemical cycles (Galloway and Cowling 2002). Within the context of debate about the onset of the Anthropocene (Lewis and Maslin 2015), there is a growing consensus that “pristine environments” probably did not exist where humans were present, because there was always some form of impact from human activities (Heckenberger et al. 2003). This perspective has been supported by archaeological research showing that ancient societies, particularly those that used agriculture, had significant impacts at both local and regional scales on terrestrial and aquatic nutrient cycles for millennia (Curtis et al. 1998; Hadley et al. 2010; Guiry et al. 2018). Evaluating the role of past human activities as drivers of ecological change has been a priority for archaeologists (Kintigh et al. 2014), but is complicated by the fact that ancient anthropogenic impacts on the environment are spatiotemporally heterogeneous and occur along a continuum of intensity (from very significant to ephemeral). For ecologists, this issue represents a source of uncertainty for retrospective studies where archaeological or paleobiological information is not available to establish when and to what extent past human activities

have impacted a particular environment or biotic community (Lyman and Cannon 2004; Holmes 2006). In contrast to the cautionary tone of the debate about the existence of “pristine” environments, our data demonstrate that, in fact, nutrient dynamics and trophic relationships in larger aquatic environments can remain unchanged despite centuries of moderate human land use.

Post-1830

Our analyses of archaeological and historical fish also show that this long-term stability was profoundly altered when Lake Ontario’s nutrient pools experienced an abrupt shift in stable nitrogen isotope composition, becoming enriched in ^{15}N throughout the pelagic ecosystem. During the early to mid-19th century, fish $\delta^{15}\text{N}$ values underwent a simultaneous, statistically significant elevation across multiple trophic levels, a difference that has been sustained throughout the 20th century and to this day (Supporting Information 5, Table S3). Atlantic salmon, for instance, which had previously had a highly conserved distribution of $\delta^{15}\text{N}$ values, show an elevation of +3‰ in well-dated mid-19th century museum specimens. Although a paucity of fish samples from the later 19th and early to mid-20th century precludes detailed comparison, these data show similar overall trends (positive shifts between intraspecific $\delta^{15}\text{N}$ means for pre-1800 and post-1990, ranging from +5.3‰ to +6.2‰, for all fish species) to those observed in Lake Ontario’s isotopic records from sediment cores (+5.5‰ to +5.6‰ for ca. 1850 to 1990s; Hodell and Schelske 1998; Fig. 2). This further indicates that the observed upward pattern in fish $\delta^{15}\text{N}$ predominately reflects a bottom-up shift in baseline $\delta^{15}\text{N}$ rather than changes to fish trophic behavior.

It is nonetheless apparent from these data that human activities have resulted in meaningful behavioral changes, including increased niche overlap between Lake Ontario’s salmonid communities. Prior to 1800, there is minimal overlap between the $\delta^{15}\text{N}$ ranges for each species, with overlaps of only 0.1‰ between Atlantic salmon and whitefishes and only 0.9‰ between Atlantic salmon and lake trout over a 600-yr period. In contrast, in the 20th century, there is substantial overlap between $\delta^{15}\text{N}$ ranges for all species in all 25-yr temporal bins (overlap ranges from 1.7‰ to 4.7‰). However, caution is required when interpreting trends in variation across time in this data set. Differences in tissue turnover rates for sample materials between pre-19th (mainly collagen from bones—slow turnover) and 20th century (mainly previously published muscle from modern fish—fast turnover) time frames make detailed analysis of these trends difficult because tissues with faster turnover rates are inherently more likely to produce greater isotopic variation reflecting seasonal or other short-term dietary aberrations. With that caveat in mind, we argue that it is likely that the greater isotopic variation in 20th century specimens reflects changes in energy pathways and foraging behavior in response to major anthropogenic environmental disruptions (e.g., impacts from invasive species,

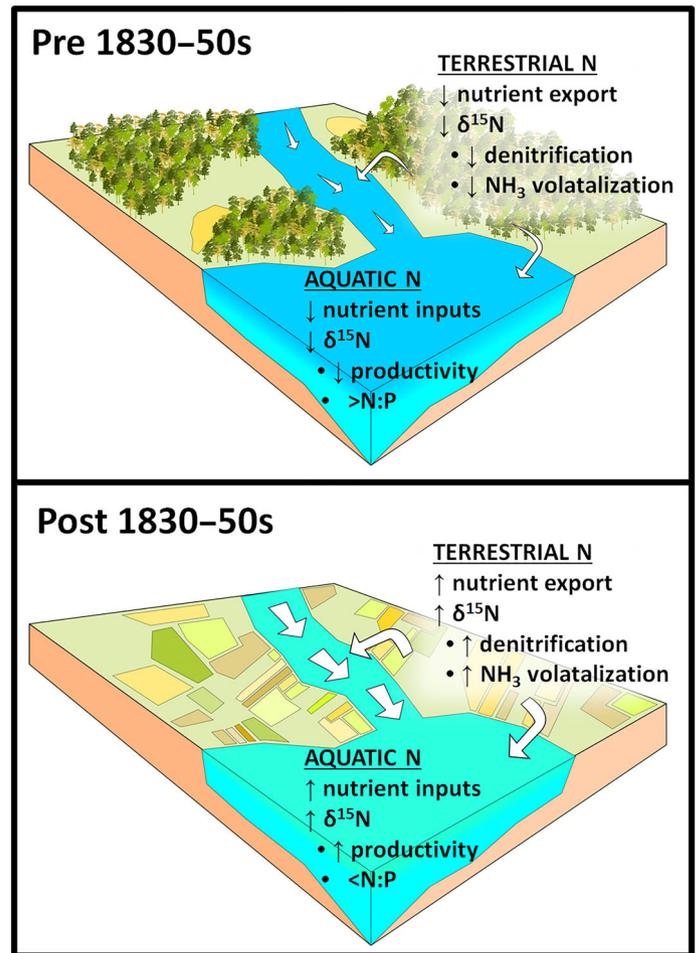


Fig. 3. Schematic diagram of nutrient regime shifts (nitrogen sources and isotopic compositions) that could be associated with the intensification of deforestation and agriculture in the Lake Ontario watershed during the early to mid-19th century.

extirpations, or over fishing; Bogue 2001; Mills et al. 2003; Colborne et al. 2016; Fera et al. 2017; Dymond et al. 2019).

The interspecific synchronicity and magnitude of this shift could have been caused by multiple related processes (Fig. 3), including changes in the nitrogen cycle (at the phytoplankton-DIN pool level) favoring nitrogen transformations that retain ^{15}N and changes in nitrogen inputs to the system that include ^{15}N -enriched nutrients (Kendall 1998; Talbot 2001; Finlay and Kendall 2007; Kendall et al. 2007). Detailed historical and archaeological dating provides a robust temporal framework to evaluate which of these variables, and therefore what type of human activity, drove this initial ecosystem-wide ^{15}N enrichment in Lake Ontario’s food web. At archaeological sites with occupation dates as late as 1832, many fish show no sign of $\delta^{15}\text{N}$ elevation for their respective taxon, whereas a museum specimen dating from no later than 1857 shows significantly elevated $\delta^{15}\text{N}$ relative to the preceding centuries. In addition, many specimens from archaeological contexts with

date ranges spanning this *terminus ante quem* (1832) and *terminus post quem* (1857) show elevated $\delta^{15}\text{N}$ values. These data therefore provide a temporal bracket for the initiation of change in Lake Ontario's nitrogen cycle, demonstrating that the shift occurred abruptly (within the context of the stability of the preceding 900 yr), within a relatively short, 15–25-yr window (1830s–1850s). This timeframe coincides precisely with the emergence of broad-scale land clearance for agriculture and timber harvesting in the Lake Ontario watershed. From the 1830s onward, especially around the middle of the century, historical analyses document how forestry activities, namely severe cutting as well as widespread burning of the remaining slash, radiated outward from the Lake Ontario waterfront to encompass much of the watershed. These forestry activities accelerated over time due to increased demand for timber (Head 1975) and cleared land, improved export capacity (canals and railroad; Lower et al. 1938; McIlwraith 1976), and growing sawmill infrastructure (Head 1975; Wood 2000).

During this period, between the 1820s and 1850s, the then-province of Upper Canada (which included the Ontario portion of the Lake Ontario watershed) had the fastest growing population in all of North America (Lewis 2001). Prior to 1830, newly settled farmers of European heritage focused on subsistence (Kelly 1973, 1975) and put considerable effort into clearing land for cultivation through cutting and burning (Ball 1979). However, relative to the mid-19th century, these early farming activities were patchy, small in scale, and diversified in purpose, with most of the land granted to settlers remaining forested (Kelly 1975). Moreover, much of the land that was cleared retained stabilizing features of the previous forest, including stumps, root mats, and stones, for up to a decade, and sometimes much longer, while under cultivation and these would have mitigated issues with soil erosion and nutrient loss (Kelly 1975; Ball 1979). During this early period, in which the farmers were themselves clearing their land, care was often taken throughout the processes, from cutting trees to sowing crops (delaying ploughing), to further minimize soil nutrient loss (Kelly 1971; Ball 1979). As farms became established, however, stumps were removed, ploughs were more widely employed to break up the soil, and more land came under cultivation. Throughout the 1830s and 1840s, the agricultural system of Upper Canada was dominated by plough-intensive (three or more times per biennial fallow) and highly profitable monocropping of wheat (Kelly 1971, 1973). The focus on a wheat-fallow-wheat agriculture system, as well as labor-saving furrow drainage and general avoidance of manure-based crop fertilization (until the 1850s) were widely blamed for rapid depletion of soil nutrients as well as erosion across the region (Kelly 1971, 1975). In this context, prior to the 1880s, when a wider appreciation developed among farmers in the Lake Ontario watershed of the risks of higher runoff-to-percolation ratios that come with overly cleared lands, farmland was subjected to a damaging seasonal cycle of spring floods, washing away

nutrient-rich organic materials, and summer droughts, parching what soil remained (Kelly 1975).

The isotopic composition of reactive forms of nitrogen in terrestrial ecosystems (i.e., soil nutrients including ammonia and nitrate), particularly those used for agriculture, is typically elevated relative to aquatic nitrogen sources (i.e., DIN) due to intense soil denitrification (Heaton 1986; Kendall 1998; Botrel et al. 2014). In this context, the most parsimonious explanation for a broad-scale isotopic shift in Lake Ontario's nitrogen cycle is that there was an increase in ^{15}N -enriched nutrient inputs coming directly from soil erosion following deforestation during the 1840s in Lake Ontario's watershed area (Fig. 3). This interpretation is supported by paleolimnological data (diatoms, sediment accumulation rates, and elemental and isotopic compositions of organic matter) from across the Lake Ontario watershed that records an uptick in nutrient loading from soil erosion runoff over the course of the mid-19th century (Hodell and Schelske 1998). The relatively early timeframe for the observed shift in fish nitrogen isotopic compositions also suggests that its cause lay in new nutrient inputs originating primarily within the local watershed area, rather than from other Great Lakes, because upstream watersheds such as Lake Erie had not been as extensively settled by this time.

While inputs of ^{15}N -enriched terrestrial nutrients, released from eroding soils across the watershed, could easily result in the observed isotopic pattern, additional related processes, occurring both on land and in the water, could further contribute to the elevation of the stable nitrogen isotopic composition of Lake Ontario's biota (Fig. 3). Higher soil erosion rates would increase mobilization of terrestrial nitrate across oxygen-poor ecotones (i.e., riparian/wetland zones), promoting bacterial denitrification, leaving the residual nutrients that are eventually delivered to Lake Ontario relatively enriched in ^{15}N (Knowles 1982; Bowden 1987; McClain et al. 2003). Increases in anthropogenic soil disturbance from farming and use of fertilizers (pushing the nitrogen cycle to be more open; Szpak 2014), as well as changes to pH in aquatic environments, may promote volatilization of ammonia before or during nutrient transport to Lake Ontario's food web (Kendall 1998). Increased phosphorous loading from soil runoff would also increase primary productivity, and the resulting bottom-up shift would have increased demand on Lake Ontario's pool of available reactive N, thereby reducing discrimination against ^{15}N during assimilation of DIN by phytoplankton (Hodell and Schelske 1998). Regardless of which of these processes were most facilitative of the nitrogen isotopic patterning in Lake Ontario's fish community, the timing (between 1832 and 1857) and abruptness of the ecosystem-wide shift indicates that the first human activity to have broad-scale impacts on nitrogenous nutrient pathways in the Great Lakes was likely deforestation.

It is also worth noting that the time period covered by this study includes significant global climatic fluctuations, including the Medieval Climate Anomaly (MCA; 950–1250 CE) and the Little Ice Age (LIA; 1300–1850 CE). Paleolimnological

studies of Lake Ontario (McFadden et al. 2005), Lake Erie (Finkelstein and Davis 2006), and other lakes in the local (Mullins et al. 2011; Paquette and Gajewski 2013; Keizer et al. 2015; Stager et al. 2017) and broader region (Laird et al. 2012) show that climatic variability associated with both the MCA and LIA did have some impact on aquatic and terrestrial environments across the Great Lakes and St. Lawrence River region. For instance, evidence from flux in biogenic silica sedimentation rates and changes in diatom (e.g., *Fragilaria* spp.) communities (Finkelstein et al. 2005; McFadden et al. 2005) demonstrates that these periods of climatic change were felt in the Lake Ontario watershed over the past 1000 yr. Climatic variability could affect Lake Ontario's nitrogen cycle and the nitrogen isotopic composition of fish through changes in adjacent wetland size (important sites for denitrification; Bowden 1987; McClain et al. 2003), terrestrial runoff impacts (Heaton 1986; Anderson and Cabana 2005), fluctuating dissolved oxygen levels (controlling nitrification and denitrification rates; Knowles 1982), and changes to productivity and thermally regulated stratification (seasonal draw-down of DIN; Hodell and Schelske 1998). Assuming that the influence of these potential sources of environmental variation did not cancel one another out, our data suggest that late Holocene climatic variation did not influence Lake Ontario's nitrogen cycle or trophic structure.

The temporal as well as taxonomical detail offered by isotopic data from archaeological fish specimens has provided a framework for higher resolution interpretation of food web dynamics as well as the timing and likely cause of the first human impacts on Lake Ontario's nitrogen cycle. While previous isotopic analyses of organic matter from sediments (Supporting Information 1 and Fig. 2; Hodell and Schelske 1998; McFadden et al. 2004) have demonstrated that Lake Ontario's nitrogenous nutrients were becoming ^{15}N enriched during the 19th century, temporal differences between data sets as well as a dearth of pre-1850 data prevented detailed analyses of the processes which drove this change in lake-wide nitrogen cycling (Supporting Information 1). In addition, although a variety of paleoenvironmental indicators (e.g., biogenic silica; Schelske et al. 1988; Schelske 1991; Schelske and Hodell 1991) have shown human activities, such as deforestation, had begun to have major nutrient (particularly P) loading impacts on the Lake Ontario watershed from the mid-19th century onward (although see McAndrews and Boyko 1972; Schelske et al. 1983), our results demonstrate that Lake Ontario's nitrogen cycle had already been substantially impacted decades earlier. Finally, by analyzing material from known taxa, we have been able to document the impact that these changes in lake-wide nutrient dynamics had on long-term food web structure.

Anthropological and ecological implications

This study is the first detailed analysis of long-term isotopic variation in freshwater fish. Results suggest that, by altering

aquatic nutrient inputs, 19th-century European forestry practices in the lower Great Lakes region, whether focused on timber extraction or producing cleared land for agriculture, left a durable isotopic signature on fish in Lake Ontario. While the findings of this research focus on the Great Lakes watershed, the implications are global; human impacts on aquatic nutrient dynamics, particularly the nitrogen cycle, may be detectable in archaeological fish remains wherever land management has extensively modified the balance of nutrient exchange between local terrestrial and aquatic landscapes. Our results are particularly germane to recent discussion in conservation, calling for increased attention to how human impacts on land may be a driver of environmental deterioration in adjacent aquatic ecosystems (Abell 2002). For this reason, our findings have significant potential to serve as a model for research that seeks to understand fundamental changes in the way past societies, through forestry and other land management practices, affected aquatic nutrient dynamics around the globe (Jenny et al. 2019).

With respect to debate on the origins of the Anthropocene, our results provide an important waypoint for the earliest lasting anthropogenic alteration of one of the world's great freshwater ecosystems. By identifying the tipping point at which human activities began to reorganize nutrient dynamics in the Great Lakes region, our results do not reveal a "golden spike" indicator for the starting point of the Anthropocene (Lewis and Maslin 2015), but, rather, provide a new signpost along the path of the global expansion of industrial impacts. These changes to Lake Ontario nonetheless represent a profound shift in the way humans have impacted the Great Lakes environment and illustrate some important anthropological and ecological considerations for how we can approach aquatic human-environment relationships in archaeology and historical ecology moving forward.

From an ecological perspective, an ecosystem-wide, bottom-up alteration of the nitrogen cycle of one of the world's largest freshwater environments over the course of only one or two decades, as shown here for Lake Ontario, demonstrates an unprecedented shift in the nature and scale of how humans cause environmental change. Understanding how humans have shaped earth's biological and physical systems remains an important archaeological and ecological challenge because finding lasting evidence of human impacts within complex regional or global systems is rare. This is particularly true for our understanding of ancient environmental nutrient dynamics, such as the cycling of nitrogen throughout an ecosystem, which, although fundamental to the success of human societies both today and in the past, preserves relatively few traces of change. Not only do our findings demonstrate how isotopic analyses of archaeological fauna can provide a clearer overview of ancient nutrient dynamics, they do so in one of the most enigmatic arenas of human-environment interaction, namely, aquatic ecosystems. Even today, in comparison with terrestrial environments, surprisingly little is known about how escalating human

exploitation of the world's aquatic environments will impact these vital ecosystems in the long term (Abell 2002). In that context, deeper retrospectives that are based on integrating results of contemporary studies with data from both historical and archaeological resources, as demonstrated by this study, have the potential to significantly enhance our understanding of the sensitivity of different environments to human disturbances (McLauchlan et al. 2013; Jeffers et al. 2015; Barak et al. 2016; Dubois et al. 2018).

From an anthropological perspective, approaching questions of human–environment interactions from the vantage point of nutrient dynamics could provide a novel outlook on the role of humans as drivers of ecological change. While the relationship between humans and their environment has traditionally been framed against a backdrop of such factors as technological change and the development of social complexity as sources of evidence for macro-scale exploitation of environmental resources (Kintigh et al. 2014), there is an increasing recognition that an understanding of how humans have influenced their landscape at a molecular level can provide powerful new lines of evidence for previously invisible but profound shifts in human–environment relations (Guiry et al. 2018). In particular, throughout most times and places in human history, the carrying capacity of local terrestrial and aquatic ecosystems, which are the environmental foundations upon which societies are sustained, has hinged on the maintenance of well-balanced biogeochemical cycles and nutrient regimes. With respect to lake and other freshwater ecosystems, which have been and continue to be of pivotal importance to many cultures, it is possible to use isotopic analyses of local fauna to look back in time and pinpoint where, when, and in that context perhaps even how, human activities reached a turning point to become dominant drivers in their ecosystem.

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Conflict of Interest

None declared.

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