J Paleolimnol (2023) 69:21–36
https://doi.org/10.1007/s10933-022-00260-x

ORIGINAL PAPER

Trends in sedimentary charcoal shapes correspond with broad-scale land-use changes: insights gained from a 300-year lake sediment record from eastern Virginia, USA

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Received: 6 December 2021 / Accepted: 26 July 2022 / Published online: 9 August 2022

Abstract Sedimentary charcoal is a useful fire proxy. Recent advances in the morphological and morphometric analyses of individual charcoal particles have enabled more nuanced paleofire interpretations. However, many uncertainties exist regarding the linkages of these particle characteristics with fuel type burned. Further, most of this proxy development research has been conducted in northern boreal biomes, which poses questions as to its universality. In this paper, we leverage a 300-year sediment record from a mill pond in southeastern Virginia, USA. Historical events and land-use changes are well constrained in Williamsburg, allowing us to compare our data with these known boundary conditions to (1) make inferences about the fuel sourcing of certain charcoal morphotypes, and (2) identify the potential controls of several charcoal morphometric characteristics. We found that the morphology and morphometry of particles in the Matoaka sediments changed in response to broad-scale shifts of historical land use and population. Prior to the American Revolutionary War (ca. 1780 CE), charcoal morphologies indicative of agricultural burning coincided with agricultural and local population expansion. Immediately preceding the American Civil War (1780–1865 CE), charcoal in Lake Matoaka recorded reforestation driven by depopulation and economic depression. After the fall of the Confederacy (ca. 1865 CE), changing charcoal morphologies reflected increased population and urban development. This increase became especially pronounced in the early 1900s, but lasted until the modern. Additionally, shifts of charcoal morphologies potentially reflect increased coal combustion. Comparison of these historical variations with morphotype shifts shows that Types M and S/B charcoal, types defined by their geometric shapes and exhibition of structure, are likely sourced from burning herbaceous fuels. However, we find that the variability of morphometric characteristics makes it unlikely that other morphologies are sourced from sole fuel types. Lastly, we propose three additional morphotypes, Types AI (angular and irregular shaped particles), E

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s10933-022-00260-x.

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(irregular, complex shaped particles), and T (triangular particles), to an existing charcoal morphological key for application in the SE USA. Overall, this research underscores the need for more work calibrating paleofire methodologies in the SE USA.

Keywords Paleofire · Proxy calibration · Charcoal morphology · Charcoal morphometrics · Fuel type

Introduction

For time periods predating instrumental data, measuring charcoal preserved in sediment records is one of the few methods for reconstructing fire histories and studying fire-climate and fire-vegetation relationships (Vachula et al. 2019; Marlon 2020). Quantifying charcoal accumulation rates has become a staple of Quaternary paleofire research (Clark 1988; Marlon et al. 2008), and this aggregate metric offers unique insight into the variability of fire activity of the past. Increasingly, new research approaches have focused on the properties of individual particles to derive additional nuance (e.g., fuel type, fire intensity) from paleofire archives (Umbanhowar and McGrath 1998; Hudspith et al. 2015; Rehn et al. 2019; Vachula et al. 2021).

The quantitative and qualitative characterization of charcoal particle shapes has facilitated interpretation of paleofire fuel type (Enache and Cumming 2006; Crawford and Belcher 2014; Leys et al. 2017; Pereboom et al. 2020; Vachula et al. 2021), an insight that was previously absent from interpretations of aggregate charcoal accumulation rates. More quantitative approaches have sought to link charcoal particle morphometrics to fuel type burned (Umbanhowar and McGrath 1998; Enache and Cumming 2006). For example, the measurement of particle aspect ratio (AR; a metric with other similar flavors including width to length (W:L) ratio, length to width (L:W) ratio and elongation) has been used to differentiate woody from non-woody fuels (Aleman et al. 2013; Leys et al. 2015, 2017; Pereboom et al. 2020; Vachula et al. 2021). Using observations of charcoal shape, texture, and porosity, several groups of researchers have developed classification systems of common charcoal morphologies with the goal of inferring fuel type burned (Enache and Cumming 2006; Jensen et al. 2007; Mustaphi and Pisaric 2014). In addition to insights gained from examining sedimentary charcoal, experimental approaches have been used to tie charcoal morphometrics and morphologies to known plant species and fuel types (Feurdean 2021; Vachula et al. 2021), as well as to characterize differences in the charcoal production rates of different fuel types (Pereboom et al. 2020; Feurdean 2021).

Geographical bias is a major shortcoming of existing charcoal classification systems. Whereas charcoal morphometric research has focused on charcoal produced in diverse biomes including temperate grasslands, tropical forest and savannah, and Arctic tundra (Aleman et al. 2013; Leys et al. 2015; Pereboom et al. 2020), research developing morphological classification systems has been limited to boreal regions of the northern hemisphere (Enache and Cumming 2006; Mustaphi and Pisaric 2014). To hypothetically use a morphological classification in a savannah ecosystem would require using a classification developed in boreal forest, regardless of the significant vegetation differences between these ecosystems. This geographical bias in paleofire research and proxy development has been noted in recent literature (Leys et al. 2018; Cheung et al. 2021; Rehn et al. 2021), highlighting the need for more complex classification systems derived from more diverse ecosystems.

Here, we present morphological and morphometric analyses of charcoal preserved in a ca. 300-year sediment record from Lake Matoaka, a former mill pond situated in a temperate forest in the southeast United States of America (USA). To our knowledge, this is the first study in the southeast USA to so comprehensively assess the morphologies and morphometric characteristics of sedimentary charcoal particles. Furthermore, we assess the applicability of these particle characteristics as proxies of broad land-use changes and subsequent shifts of fuel type in this geographic location.

Study site

Our study focuses on Lake Matoaka, a former millpond in Williamsburg, Virginia, USA (Fig. 1) and within the Atlantic Coastal Plain. Lake Matoaka was dammed as a millpond in the early 1700s CE, and as such, is confined within several narrow valleys and fed by four perennial streams. Today, the lake is owned by the College of William & Mary, whose main campus borders Matoaka to the east and is otherwise surrounded by the College Woods, a forest

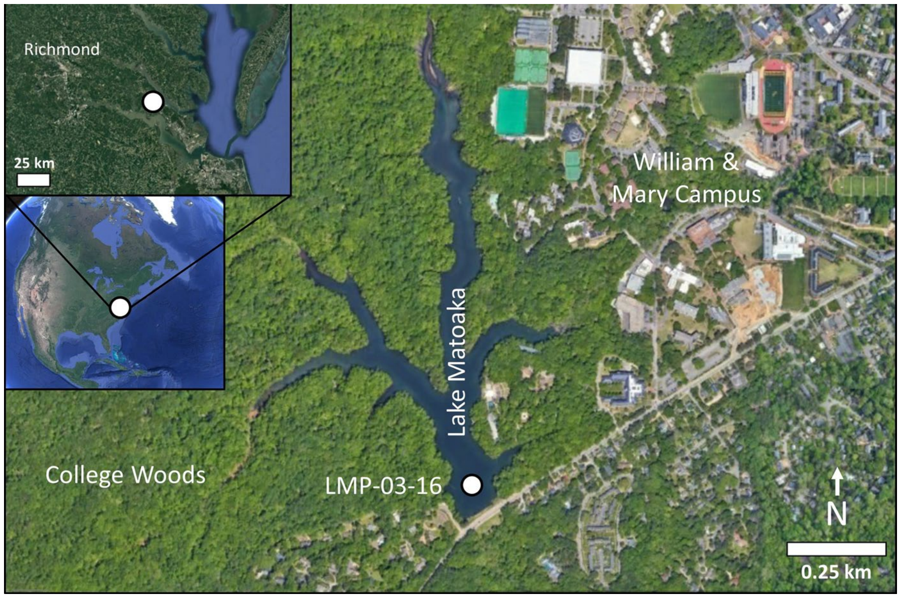

Fig. 1 Map of the study site (Imagery from Google Earth). Inset shows proximity to Richmond metropolitan area and location of Williamsburg within eastern Virginia

stand dominated by oak, hickory, birch, poplar, and maple (Monette and Ware 1983; Cyrus 2016). The watershed of Lake Matoaka (6 km^2) is characterized by ~ 60% forest and ~ 40% residential land (Evans and Packard 2007). Overall, the lake is relatively small, with a present day area of 0.17 km^2 and a maximum depth of 4.8 m in the main channel.

The sediments in Lake Matoaka span the last ~ 320 years and record the history of the region's socio-economic development as well as its conflicts. Together, these historical events have driven dynamic changes in local land use from the colonial period through industrialization which reflect a broader national trend of agricultural expansion followed by abandonment and reforestation (Balascio et al. 2019). In 1699, Williamsburg became the colonial capital of Virginia. As the capital, Williamsburg experienced a boom of economic and development activity (Morgan 2004). The damming of Lake Matoaka reflected a broader trend of agricultural expansion and the creation of mill dams (Stephenson 1947). Intensive agriculture, deforestation, and industry continued until the American Revolutionary War (CE 1775–1783). The capital was moved westward to Richmond in 1780 CE due to the approach of British forces, taking nearly half of Williamsburg's population with it (Morgan 2004). This, along with post-War depression, economically deflated Williamsburg for the next several decades, allowing for some reforestation (Handler and Gable 1997; Balascio et al. 2019). Following the American Civil War (CE 1861–1865), Williamsburg's population increased by as much as 80% by the early 1900s (Morgan 2004), which was matched by a long-term increase in development. In 1920, restorative reconstruction of Colonial Williamsburg began and Lake Matoaka and its surrounding lands were purchased by the College of William & Mary.

Materials and methods

Lake Matoaka sediments

In March 2016, a 148-cm sediment core (LMP-03–16; (37° 15.878′ N, 76° 43.386′ W; 7 m asl) was collected from Lake Matoaka and an age-depth model was developed for this core using ^{210}Pb, ^{137}Cs, and $^{206}Pb/^{207}Pb$ (Balascio et al. 2019) (ESM1 Fig. S1). For complete sedimentary descriptions and age model development, see Balascio et al. (2019). The age-depth model shows that the average sedimentation rate in the lake was 0.4 cm $year^{-1}$. The basal sediments of the core are sandy peat indicating wetland environment, with lacustrine sediments beginning at 126 cm depth, where sandy, organic-rich peat transitions to a less-dense light brown clayey-silt unit with some coarse intervals and terrestrial macrofossils throughout (Balascio et al. 2019).

Charcoal morphometric and morphological analysis

Thirty-six contiguous subsamples were taken from the lacustrine units of core LMP-03-16 (4-cm resolution from 126 to 55 cm and 2.5-cm resolution from 55 to 0 cm (resolution change was made to account for change in sedimentation rate)). Subsamples were originally processed for spheroidal carbonaceous particles (SCP) analysis (Rose 1994; Cahoon 2019). Dry sediment was digested with H_2O_2 and strong acids (HNO_3, HF, and HCl) to eliminate organic matter, siliciclastic clays, heavy minerals and rock fragments. Samples were centrifuged at 3000 RPM for 5 min after each digest (except HF, which was allowed to settle for 24 h) to remove the supernatant. Digested resides (200 µl) were mounted onto slides with Norland optical adhesive and a cover slip. Some research shows that chemical treatments like those we undertook could digest charcoal particles that are not fully charred and/or were formed in low intensity, low temperature fires (Schlachter and Horn 2010; Constantine IV and Mooney 2021), so it is possible the charcoal we analyzed tended to be formed in more intense, higher temperature fires. However, the chemical digestions are analogous to those commonly used to analyze charcoal on slides (Daniau et al. 2007; Genet et al. 2021). Additionally, there was some worry that the physical processing and centrifuging might affect our charcoal measurements. We tested the potential impact of this physical processing and found that although there were some differences between the samples subjected the physical processing (relative to control samples which were not), the differences were within the likely natural range of charcoal concentrations between samples and did not exhibit a consistent pattern suggesting breakage of particles by the physical processing (ESM1 Table S1).

Using a Leica DM750 microscope with a ICC50 W attachment and Leica Application Suite V4.12, slides were photographed at 30× magnification (with the exception of one sample whose overall particle assemblage was small enough to require 50×). In each slide, a random set of 100 charcoal particles was identified. CharTool, an open-source ImageJ macro (Snitker 2020), was used to measure charcoal particle morphometric characteristics and record user-defined morphologies. We used CharTool's scale feature to ensure accurate morphometric measurements. Charcoal morphotypes were classified using CharTool's modified form of the Enache and Cumming (2006) system (Fig. 2; black). Although CharTool collects 21 morphometrics, we present only the morphometric variables of height (the height of the smallest rectangle enclosing the selected pixels), area (the area of the selected pixels in the units assigned to the software), circularity ($4\pi \times \text{area} \div \text{perimeter}^2$, wherein perimeter is the length of the outside boundary of selected pixels in µm), aspect ratio (AR; the major axis of the best-fitting ellipse around the selected pixels divided by the secondary axis of said ellipse), roundness ($4 \times \text{area} \div \left(\pi \times \text{major axis}^2\right)$), and rectangularity ($\text{perimeter} \div ((\text{width} + \text{height}) \times 2)$).

Aggregate charcoal analysis

Dry sediment was weighed, utilizing bulk density measurements (Balascio et al. 2019), to approximate 1 cm^3 of wet sediment. Following Vachula et al. (2018), the samples were immersed in 12 ml of 50% sodium metaphosphate and 50% bleach for 48 h and then washed over nested 125 and 63 µm sieves. These sieve sizes were chosen to characterize local and regional signals of fire history, respectively. Sieved samples were transferred to a gridded petri dish and charcoal particles were enumerated using a binocular dissection microscope. Charcoal counts were converted to charcoal accumulation rate (CHAR) using the following equation:

$$\text{CHAR} = \frac{\text{number of particles}}{\text{volume of sample}} \times \frac{(\text{Bottom depth} - \text{top depth})}{(\text{Top age} - \text{bottom age})}$$

Grain size analysis

Samples were pretreated with 10 ml of 30% hydrogen peroxide for 48 h to remove organic matter, and for 24 h with 10 ml of hexametaphosphate to disaggregate particles before analysis on a Beckman Coulter LS13320 laser diffraction particle size analyzer (Balascio et al. 2019).

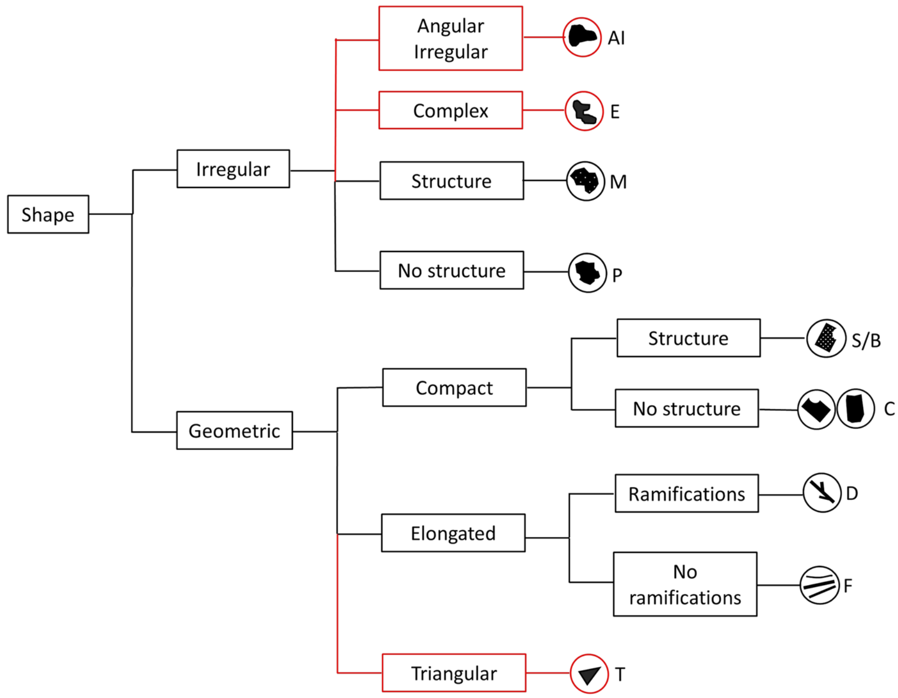

Fig. 2 Schematic of the Enache and Cumming (2006) charcoal morphotype classification system (black) and our proposed amendments for application in the SE USA (red)

Statistical analyses

We used Pearson's correlation coefficients (r) and associated null hypothesis testing (p) to quantify visual relationships between variables.

Results

Charcoal morphometric and morphological analysis

In total, we identified, classified, and measured 3,598 charcoal particles preserved in Lake Matoaka sediments. The Type C morphotype was dominant (54%), with Types P (18%), S/B (11%), F (8%), M (7%), and D (2%) constituting much smaller proportions of the charcoal assemblages (Fig. 3A).

Types F, S/B and M had the largest particle areas; while types C, and D had smaller area values (Fig. 3B). Types C, P, S/B and M broadly had greater roundness values than Types F and D. Types F, P and S/B had similar circularity values, while M and D had lower circularity values (Fig. 3C). Type C had the highest circularity values. Types C, P, S/B, and M had low aspect ratios (AR), while types F and D had larger and more variable AR values. Measured particles become more rectangular as their rectangularity values approach 1.0; approaching 0.0 such particles become more circular and surpassing 1.0 they become more irregular (Snitker 2020). Types C, F and P were the most rectangular, while types S/B, M and D were more irregular.

The most abundant morphotype in every sample, type C, had relatively constant percentages from 1720–1813, 1813–1884, and 1884–2012 (Fig. 4). Type D percentages were relatively constant from 1720 to 1813, showing one spike of 8% 1884, and were only slightly more variable from 1884 to 2012. Type F percentages were relatively constant from 1720–1813, were higher but more variable from 1813 to 1884, and were highly variable with a slight increase from 1884 to 2012. M percentages were variable from 1720 to 1813, exhibited low variability from 1813 to 1884 and were higher on average from 1884 to 2012. Type P percentages were relatively constant from 1720 to 1813, increased slightly from 1813 to 1884, and were highly variable from 1894 to 2012. Type S/B particles exhibited the highest average composition abundance from 1720 to 1813, decreased in average percentage slightly from 1813 to 1884, and composed significantly less of each sample on average from 1884 to 2012 (Fig. 4).

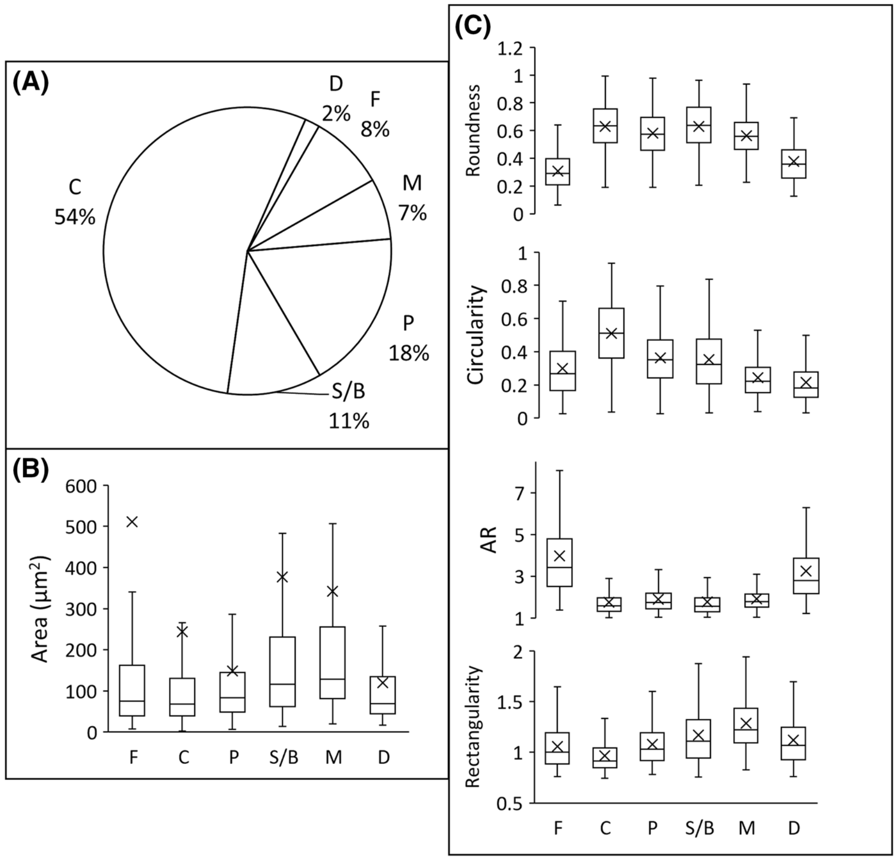

Fig. 3 Morphologies and morphometric measurements of charcoal from Lake Matoaka sediments. The charcoal particles identified were dominated by the Type C morphotype but there was a diversity of morphotypes present in the sediments (**A**). The sizes of the morphotypes varied but were comparable (**B**). The morphometric characteristics (note: these indices are unitless) of each morphotype varied considerably (**C**). These data are provided in ESM1 Table S2

Charcoal accumulation rates (CHAR) of particles > 63 µm and > 125 µm in size covaried through time (correlation coefficient = 0.65, $p < 0.01$). CHARs of the > 63 µm fraction were variable from 1720 to 1813, generally decreased from 1813 to 1884, and then increased from 1884 to present. CHARs of the > 125 µm fraction were generally comparable from 1720 to 1813 and 1813–1884, but increased from 1884 to present (Fig. 4).

We observed that the slide mounting adhesive appeared to fracture charcoal particles in situ during the curing process (Fig. 5). Additionally, a number of triangular particles were observed throughout the record and were classified as Type C (compact, geometric, structureless particles), as per Enache and Cumming (2006).

Discussion

Historical variations in charcoal accumulation, morphology, and morphometry

The morphology and morphometry of particles varied markedly over time. The charcoal assemblages preserved in the Matoaka sediments can be divided into three time periods by similar morphological and morphometric characteristics. From ca. 1720 to 1815 CE, Types S/B, M, F, and D are quite variable, whereas P and C are relatively stable at approximately 20 and 50 percent of each sample, respectively. From 1815 to 1885 CE, Types C and P are stable, whereas Types D and F exhibit marked fluctuations. During this time

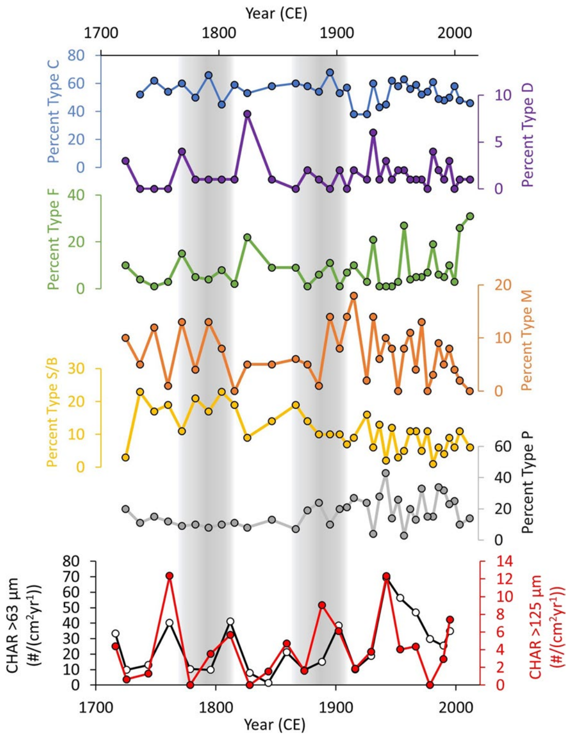

Fig. 4 Historical variations of charcoal accumulation and morphological assemblages in Lake Matoaka correspond with historical population changes and subsequent shifts in land use. Gray shading depicts transitions of morphological assemblages. These transitions co-occur with historical population changes and broad scale regime shifts of land use in association with the American Revolutionary War and the movement of the capital to Richmond (ca. CE 1775–1810) as well as the post- Civil War increases of population in Williamsburg (ca. CE 1865–1900). These data are provided in Table S3

period, the abundance of Types M and S/B generally decreased. Charcoal deposited during this period also exhibits relatively high average aspect ratio (AR) values (Fig. 6). From 1885 to present, Types C and P displayed more variability than in earlier periods, as did Types M, F, and D. In recent decades, Types M, P, and F increased substantially, whereas Type S/B decreased. These morphological changes are further reflected in the morphometric values of these charcoal particles. For example, the average height, rectangularity and roundness of particles were relatively low prior to ca. 1900 CE, after which they both increased substantially. Much research has linked these morphotypes and morphometrics with fuel sources, so the shifts in charcoal assemblages evident between these three periods (1720–1815, 1815–1885, and 1885-present) are likely indicative of changes in fuel type burned (Umbanhowar and McGrath 1998; Enache and Cumming 2006; Jensen et al. 2007; Crawford and Belcher 2014; Mustaphi and Pisaric 2014). Notably, the peak in charcoal accumulation centered at 1761 CE could correspond to increased fire activity associated with Williamsburg's role in the American Revolution. The opening of the James Anderson Armory ca. 1765 CE and the charging of this armory with the task of equipping the American troops resulted in a massive use of fuels and forges (Jaworski 2017; Cahoon 2019).

We infer that historical land use and population variations caused the broad-scale shifts of charcoal assemblages that we observe in the Lake Matoaka sediments. The first shift occurs ca. 1800 CE, which generally coincides with the movement of the capital from Williamsburg, subsequent depopulation, and economic depression that occurred following the American Revolutionary War (Fig. 4). Similarly, the second shift of morphology assemblages occurred ca. 1880 CE, which corresponds with the increased population and development in Williamsburg following the American Civil War (Fig. 4). In addition to anthropogenically-driven shifts in charcoal morphology and morphometry, natural wildfire likely had a role in altering the charcoal signal and assemblages in the Lake Matoaka sediments. However, differentiating natural and human-caused changes in paleofire archives is notoriously difficult (Roos et al. 2019; Marlon 2020). Distinguishing whether charcoal is sourced locally or regionally is similarly challenging (Vachula 2021), although charcoal is a primarily local signal (Clark 1988; Higuera et al. 2011). The size of the charcoal particles identified for morphological and morphometric analyses varied but tended to be ~ 100 µm^2 (the equivalent of ~ 10 µm if sieved). Charcoal sieved to quantify charcoal accumulation rates was greater, with size fractions of > 63 and > 125 µm. Charcoal source area typically increases with smaller particle size at single sites (Vachula 2021), so it is likely our sieved charcoal records slightly different spatial scopes of fire history. However due to the spatially autocorrelated nature of fire as well as the primarily localized signal of any given size fraction of sedimentary charcoal (Clark 1988; Higuera et al. 2011; Vachula and Richter 2018; Vachula et al. 2018), it is likely our three size fractions provide generally compatible perspectives

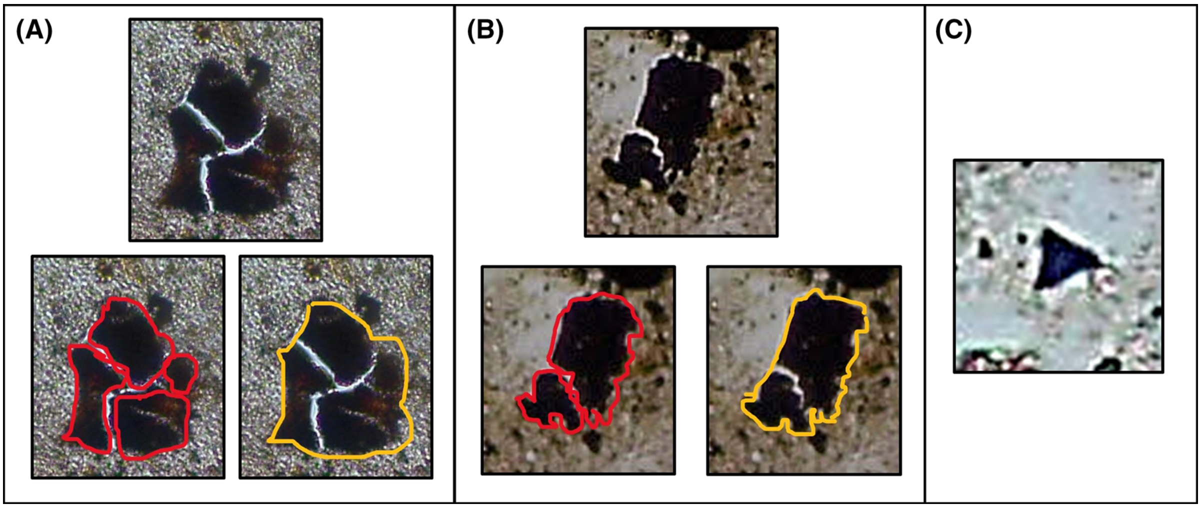

Fig. 5 Photographs of charcoal particles preserved in the Lake Matoaka sediments. We observed that in several cases (**A** 0–2.5 cm depth (horizontal field of view = 100 µm); **B** 27.5–30 cm depth (horizontal field of view = 40 µm), the optical adhesive appeared to have caused fracturing of the particles. These fractured particles provide an opportunity to assess how particle breakage might affect morphotype classifications. Additionally, we observed multiple triangular charcoal morphotypes (e.g., panel **C** 52.5–55 cm (horizontal field of view = 20 µm)), which we propose warrant an additional morphotype category when analyzing sedimentary charcoal in the SE USA (Fig. 2)

on regional fire history surrounding Lake Matoaka on the order of 10–100 s of km (Vachula 2021). Given that Lake Matoaka is centrally located within the Williamsburg metropolitan area, and the historical population variations of Williamsburg generally reflect those of the broader regional surroundings, so we conclude that human activities likely dominate the paleofire signals within the Lake Matoaka sediments.

Linking known historical land-use changes to charcoal particle assemblages preserved in Lake Matoaka

The creation of the lake and deposition of its earliest sediments ca. 1700 CE coincides with a period of agricultural expansion and development, and a population increase to 2000 (Morgan 2004). This expansion lasted until the depopulation of Williamsburg in response to the American Revolutionary War ca. 1780 CE (Morgan 2004). As such, we might expect charcoal indicative of consistent agricultural burning during this period. Indeed, charcoal assemblages during this period are marked by elevated proportions of Type M and S/B charcoal, morphologies that are distinguished by their visible cellular structures (Enache and Cumming 2006). These cellular structures are typically indicative of grass or leaf stoma (Mustaphi and Pisaric 2014), which supports our inference of sourcing from agricultural burning (Fig. 7).

From ca. 1780 CE to the American Civil War (beginning in 1865 CE), reforestation is likely to have occurred during a period of depopulation and economic depression (Handler and Gable 1997; Balascio et al. 2019). During this period, we might expect agricultural burning to decrease and natural wildfire to dominate biomass burning signals. Overall, we observe from charcoal accumulation rates (CHAR) that there was a general decrease in fire activity during this period. Further, we observe that the relative proportions of Types M and S/B (indicative of agricultural burning prior to this period), generally decrease from ca. 1780 to 1865 CE. At the same time, elongate Types D and F increase in their relative abundance. Although Types D and F have been tied to leaf vascular structures and grasses, they have also been suggested to be produced from wood and twigs (Enache and Cumming 2006; Mustaphi and Pisaric 2014), which would support our inference of changing fuel sources. In contrast, the average aspect ratio of particles during this period was relatively high, suggesting increased non-woody fuels (Umbanhowar and McGrath 1998; Crawford and Belcher 2014;

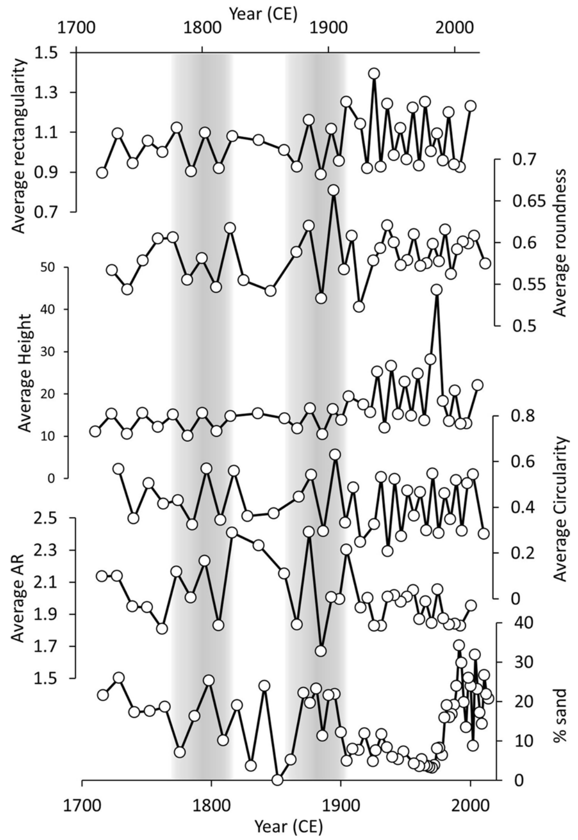

Fig. 6 Historical variations of charcoal morphometric values in Lake Matoaka. Gray shading depicts transitions likely associated with historical population changes and subsequent shifts in land use in association with the American Revolutionary War and the movement of the capital to Richmond (ca. CE 1775–1810) as well as the post-Civil War increases of population in Williamsburg (ca. CE 1865–1900)

Pereboom et al. 2020; Vachula et al. 2021). Altogether, it therefore seems that from ca. 1780 to 1865 CE, overall fire activity decreased, but the fire activity which persisted was composed of both natural forest fires and agricultural burning (Fig. 7).

Following the Civil War (from ca. 1865 CE to the beginning of the twentieth century), Williamsburg's population increased and urban development surged (Morgan 2004), a trend that continued into the twentieth century. Therefore, following the Civil War, we might expect a relative decrease in agricultural burning and subsequent charcoal morphotypes. Indeed, we observe a decrease in the relative abundance of Type S/B, which we previously linked with agricultural burning (Fig. 7). Similarly, we observe increases of particle rectangularity, and the relative abundance of Type F, which could reflect increases of woody fuels (Enache and Cumming 2006; Mustaphi and Pisaric 2014). However, we observe a relative increase of Types M and P charcoal, which do not align with these inferences, given previously published guidance. Given that the relative increase of Type P charcoal is limited to this modern period, we infer that these particles could be reflective of coal fuel sources or other anthropogenic burning associated with industrialization and increasing development (Blarquez et al. 2018; Cahoon 2019). Similarly, initial work describing Type P noted its powdery composition (Enache and Cumming 2006), suggesting potential sourcing from coal and/or higher combustion temperature (Masiello 2004; Enache and Cumming 2006; Mustaphi and Pisaric 2014). Alternatively, it is also possible that Types M and P charcoal are derived from leaves and agricultural burning, but that this sourcing has not be previously noted in the published literature.

Altogether, these historical land-use changes appear to correspond with shifts in the charcoal assemblages preserved in Lake Matoaka. However, many uncertainties exist regarding the source fuels of charcoal morphotypes (Enache and Cumming 2006; Jensen et al. 2007; Mustaphi and Pisaric 2014), as well as the reliability of morphometric data as indicators of changing fuels (Umbanhowar and McGrath 1998; Crawford and Belcher 2014; Vachula et al. 2021).

Connecting charcoal particle characteristics to fuel source: insights gained from Lake Matoaka

Type C was the dominant morphotype in the Lake Matoaka sediment record, encompassing more than half of all particles identified. As defined by Enache and Cumming (2006), Type C consists of compact, geometric charcoal with no visible structure, and several lines of evidence suggest that Type C charcoal is sourced from wood fuels (Umbanhowar and McGrath 1998; Enache and Cumming 2006). However, despite stark land use changes and shifts in the dominance of wood fuels, the relative proportion of Type C charcoal in the Lake Matoaka sediments remained steady. Notably, Type C also dominated the charcoal assemblages of the boreal forest site presented by Enache and Cumming (2006). We infer that the broad

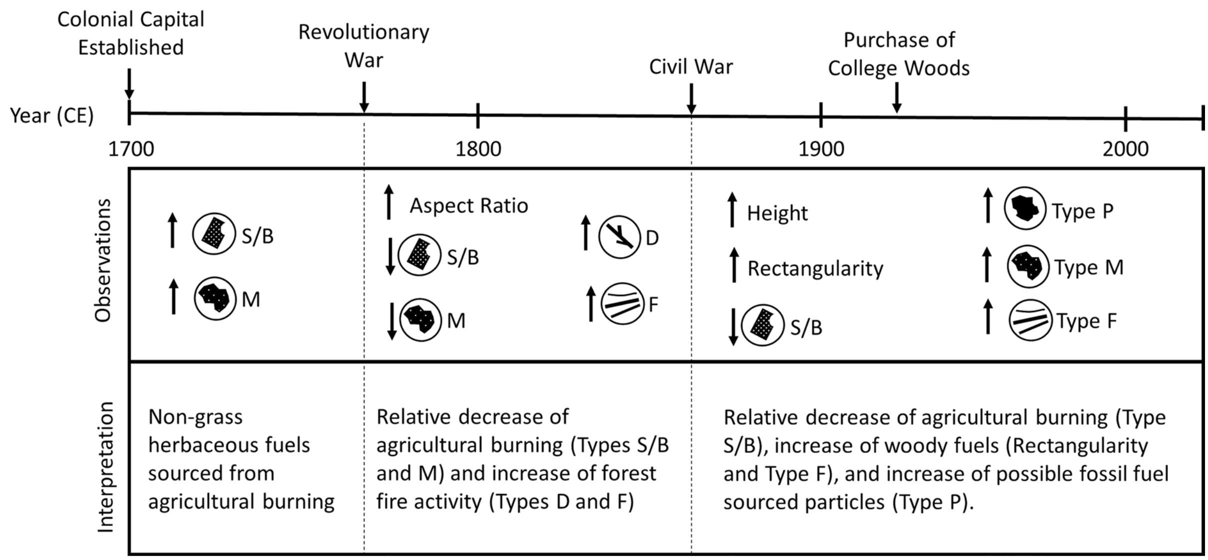

Fig. 7 Conceptual diagram depicting general observations of historical changes in the charcoal morphologies and morphometrics preserved in Lake Matoaka. Our interpretation of these changes is also shown

definition of Type C, rather than a veritable dominance of wood fuels, can explain this discrepancy in the Matoaka sediments.

The least abundant morphotype in the Matoaka sediments was Type D, which is characterized by elongated particles with ramifications (Fig. 3). The relative dearth of this morphotype could either relate to the charcoal yield of the fuel source generating that morphology, or the lack of abundance of that fuel source in this region over the past 300 years. Jensen et al. (2007) identified similar elongated, branching charcoal particles to be produced from the veins of deciduous leaves, specifically oak (2007). Deciduous trees are common in the forested Lake Matoaka catchment and the broader region (Terlizzi 2021), despite their apparent absence in the charcoal record. This disconnect suggests that either burning deciduous leaves yield less charcoal than other fuels, or that the charcoal they produce is more fragile and breaks down into smaller fragments, no longer recognizable as Type D. Experimental burn data show that deciduous leaves yield more charcoal mass than various woody fuel types (Feurdean 2021), casting doubt on this explanation. Therefore, the fragility of Type D particles could explain the lack of these morphotypes, in line with previous study (Umbanhowar and McGrath 1998; Enache and Cumming 2006). Alternatively, it is also possible that the triangular particles we observed were produced from deciduous tree leaves but were not coded as such due to limitations of the classification key.

The association of Types M and S/B charcoal with known periods of agricultural expansion and development, in combination with their morphometric characteristics, indicate that these morphotypes are likely sourced from burning herbaceous fuels. The stoma and/or fibrous structures inherent to these morphotypes have been previously linked to grass or leaf fuels (Mustaphi and Pisaric 2014). However, we observed that Types M and S/B morphotypes have among the lowest aspect ratios (AR), which indicates these particles are unlikely to be sourced from grass fuels (Umbanhowar and McGrath 1998; Crawford and Belcher 2014; Vachula et al. 2021). However, the correspondence of fluctuations in relative abundance of Types M and S/B with known periods of variations in agricultural intensity (and subsequent biomass burning) indicate that these morphotypes are likely sourced from herbaceous fuels produced by agricultural burning.

The precipitous increase of Type P charcoal following the Civil War (ca. 1865 CE) and lasting until the modern, in tandem with subsequent development and population increases beginning in the twentieth century, suggest that this charcoal morphotype is sourced from fossil fuels. The distinct increase in the relative abundance of Type P charcoal generally corresponds with known increases of fossil fuel consumption, which are also recorded in the Lake Matoaka sediments (Balascio et al. 2019). Further, initial work describing Type P noted its powdery composition (Enache and Cumming 2006), which supports our interpretation that this morphotype is sourced from coal and/or higher combustion temperature.

Although elongate charcoal morphologies are posited to distinguish fuel types (Enache and Cumming 2006; Mustaphi and Pisaric 2014), a comparison of our morphological determinations with morphometric data indicates that qualitative morphologies may be insufficient to differentiate fuel types. The aspect ratio (AR) of charcoal particles has been shown to be a reliable proxy of grass versus woody fuel sources (Umbanhowar and McGrath 1998; Aleman et al. 2013; Crawford and Belcher 2014; Vachula et al. 2021). As expected, the two elongated morphotypes, Types F and D, had the greatest aspect ratios (Fig. 3). However, they also had the two greatest ranges of AR values (Fig. 3), suggesting that a significant proportion of this morphotype is composed of elongated particles that are not metrically elongated, despite a qualitatively elongate appearance. If this is the case, it is likely that these morphotypes are derived from both grass and woody components, a possibility that was noted by Enache and Cumming (2006). In this way, qualitative morphologies may be unable to distinguish fuel types in the same way that quantitative morphometrics are able. Further, the mixture of charcoal morphotypes derived from fuels that are less readily distinguishable may convolute the quantitative signals (e.g., AR values) that are typically coherent when comparing more distinct fuels.

The variability of morphometric characteristics within classified morphotypes of charcoal preserved within the Lake Matoaka sediments suggests that morphotypes are unlikely to be sourced from sole fuels. Although several distinct differences exist between the means and distributions of morphometric data of charcoal morphotypes (Fig. 3C), the considerable variability and ranges of these morphometrics underscore the potential variability of the shapes that could qualify as various morphotypes. Generally speaking, morphometric data tends to be specific between fuel types (Umbanhowar and McGrath 1998; Crawford and Belcher 2014; Vachula et al. 2021), a reality not exhibited by the morphometric characteristics of morphotypes in Lake Matoaka. As such, we infer that the morphotypes of charcoal in Lake Matoaka have multiple possible fuel sources. Our approach combining morphometric data with morphological classification has important implications for future paleofire research; the interpretation of morphotypes must attribute fuel sources conservatively. Morphology alone may not be a definitive indicator of fuel source because the morphologies of charcoal produced from different plants can overlap, and so morphologies should be employed with other analyses.

The potential effects of experimental methodology on charcoal particle characteristics

The fracture of charcoal particles that we observed in our slides is a fortuitous opportunity to investigate the possibility of charcoal particles changing morphotype when fractured. Many charcoal particles within our record appeared to be broken from the same larger particle (Fig. 6). In most instances, the morphologies of the new, smaller particles were not the same as that of the original, unified particle (Fig. 6). We infer that this systemic fracturing of particles may be due to thermal contraction of the slide mounting resin (Norland Optical Adhesive; 1.5% linear shrinkage). Although this contraction and subsequent fracture of charcoal particles is noteworthy and should be considered with caution in future research, it also sheds light on charcoal particle breakage as a taphonomical process. For example, we show that two Type C morphotypes could produce a mixture of morphotypes when broken (Fig. 6), a possibility that was also noted in earlier research (Enache and Cumming 2006). The possibility that smaller charcoal fragments of one morphology are actually representative of the breakdown of a larger particle of another morphology poses an inherent problem for this fuel proxy. Further empirical work is needed to test the degree to which breakage via transport and weathering might affect the utility and reliability of morphological groups and their links to fuel types.

A new classification scheme for the southeast USA

Our finding that the majority of charcoal particles (54%) preserved in Lake Matoaka belong to the same morphotype (Type C) indicates that either (1) the majority of charcoal particles are derived from the same fuel source and so exhibit the same morphology, or (2) that the morphological key we used requires more detailed morphotype options to be applied to the SE USA (Enache and Cumming 2006). Informed by our observations of the Type C particles in the sediments, we infer that this category is too broad for application to the SE USA. Type C had the greatest range in circularity values in our dataset, despite the angularity inherent to its definition (Fig. 3C). This disconnect is likely due to the broad definition of the Type C morphotype, which includes both angular and angular-irregular particles not exhibiting structural characteristics. Additionally, irregular particles in the Enache and Cumming (2006) taxonomy are fairly compact, leaving a gap in our ability to label the numerous irregular particles with branching, protruding, or otherwise non-compact structure; irregular particles with these features have been characterized in previous literature (Mustaphi and Pisaric 2014).

To prevent broad categorization of particles with distinctly different morphological properties, and therefore possibly varied fuel sources, we propose three new morphotypes for our key in the SE USA. In light of the problems we identified with the applicability of the Enache and Cumming (2006) taxonomy, we propose that two morphotypes be used to allow more nuanced characterizations of geometric and irregular charcoal particles. These morphotypes, Types AI (angular-irregular shaped particles) and E (irregular, complex shaped particles) would be useful additions to the Enache and Cumming (2006) key in this region (Fig. 2; red). Additionally, we propose Type T, triangular particles, to be added to the Enache and Cumming (2006) key for use in the SE USA (Fig. 2; red). In our survey of the charcoal particles preserved in Lake Matoaka, we noted abundant triangular particles. These particles were most often classified as geometric and without structure (Type C), but occasionally as irregular if their sides were significantly curved or sloped (Type P). Inclusion of the Type T morphotype in future study of charcoal particles in the SE USA could further subdivide the dominant Type C group, leading to counts driven by particle abundance instead of aggregation under a 'best fit' definition. Figure 8 demonstrates the success of this subdivision, as our proposed classification was able to differentiate particles previously classified as Type C. Although it is difficult to determine the fuel source of these triangular Type T particles, we infer that they may be produced from the terminal ends of conifer needles. It is also possible that the Type T particles were produced from deciduous tree leaves and/or fern leaves, which would reconcile the apparent absence of Type D charcoal derived from the veins of deciduous leaves in the Lake Matoaka sediment record.

Application of our proposed revisions to the Enache and Cumming (2006) key (Fig. 2) more reliably differentiate and characterize the diversity of charcoal assemblages preserved in Lake Matoaka. Using our revised key, we revisited three Lake Matotaka sediment samples and reclassified the charcoal morphotype assemblages. Our revised key was successful in its ability to better characterize the diversity of charcoal morphotypes (Fig. 8). Importantly, each of these samples falls within the three broad periods of charcoal variations in the Lake Matoaka sediment record (Fig. 7), showing that this revised key is useful across time periods with varying fuel types. Altogether, these data serve as a proof of concept that our revised key will be readily useful and applicable for paleofire records of the SE USA.

Our observation of a morphological key with an imperfect application to a sedimentary charcoal record highlights a broader issue of regional specificity in paleofire research. Charcoal morphological keys must be used with caution when applied outside the biome of their development. Transferring morphological keys between sites assumes that all morphotypes encountered will be represented within the key. To our knowledge, no globally applicable taxonomy has been developed. Rather, region-specific keys and studies have been undertaken with varying degrees of implied universality. For example, (Mustaphi and Pisaric 2014) developed an expansion of the Enache and Cumming (2006) key using sedimentary charcoal in a Douglas fir dominated forest, suggesting that a degree of modification dependent on locally observed morphotypes would allow the application of their key in other regions. Whereas the bulk of charcoal morphological keys are based on records from forested regions (Enache and Cumming 2006; Jensen et al.

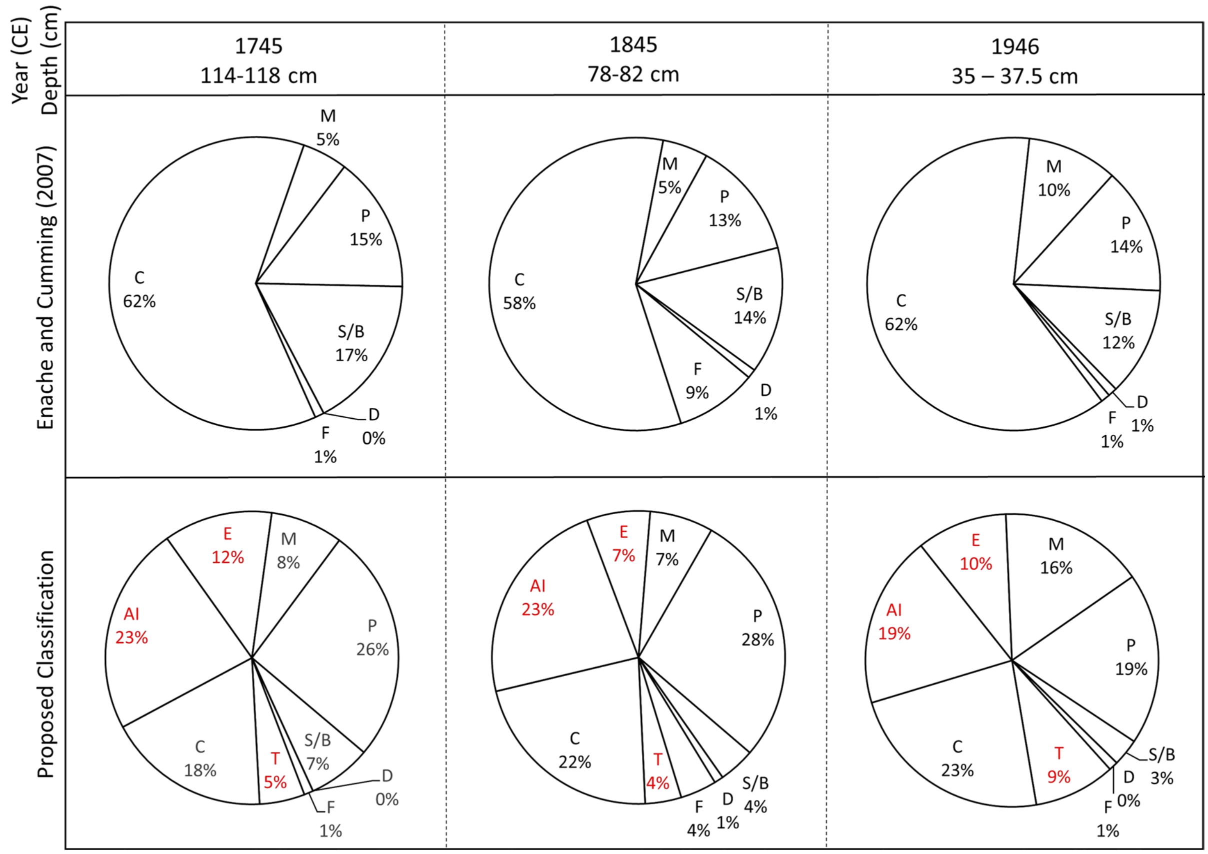

Fig. 8 Application of our proposed revision of the (Enache and Cumming 2006) charcoal morphotype classification system. For three samples of the Lake Matoaka sediments, we applied our revised classification (Fig. 2). Results show that the inclusion of new morphotypes (red; Types T, AI, and E) successfully differentiates variability within the more general morphotypes of the Enache and Cumming (2006) classification system (e.g., Type C)

2007; Mustaphi and Pisaric 2014), many other studies have focused on charcoal in grassland dominated regions without proposing a morphological key (Aleman et al. 2013; Leys et al. 2015, 2017). More study is needed to create a strong body of morphological data within each region of interest. Significant pitfalls can exist for fuel type interpretations of charcoal particles when applying a key in a region outside of its intended use (Rehn et al. 2021), highlighting the need for recognition of potential mismatch and subsequent caution. For example, Cheung et al. (2021) successfully use the Mustaphi and Pisaric (2014) key to analyze charcoal from an Indonesian cropland catchment with some shrubland and open forest, but noted that the key application suggested some fuel sources which did not exist near the study site. The relative diversity of the Mustaphi and Pisaric (2014) key suggests it may indeed be more widely applicable; even so, further work in additional contexts is needed to fully test whether morphological keys are appropriate and effective in all biomes of the world.

In addition to potential vegetation differences, the size range of charcoal analyzed in the development of a key must be considered in its transfer to a new record or location. The bulk of charcoal morphological literature focuses on macroscopic charcoal, a size class of charcoal typically encompassing charcoal greater than 125 µm in size (Vachula 2019). In this study we did not use sieves when isolating charcoal for morphometric analysis and morphogical classification, but the majority of charcoal particles were smaller than 150 µm in diameter. This is an important caveat of our analysis; although we focused on particles typically defined as microscopic charcoal

(Vachula 2019), the key we employed was developed for macroscopic charcoal. One potential manifestation of this mismatch is the relative paucity of Type S/B in the Lake Matoaka sediments, as some of the structures inherent to their definition may have been too large to be preserved in the size class of charcoal we identified. Alternatively, by not using a sieve, we may have been more accurately characterizing the sizes of charcoal particles in the SE USA, which may trend smaller than the macroscopic particles dominant in northern boreal forests.

More broadly, our analysis highlights that more work is needed to establish paleofire methodologies in the SE USA. There has been little paleofire research in the SE USA relative to other regions of the USA, such as the West (Power et al. 2008; Marlon et al. 2012). This deficiency is problematic in light of the large human populations and subsequent large areas of wildland-urban interface in the SE USA (Radeloff et al. 2005; Theobald and Romme 2007), which are at risk to fire (Cohen 2000). However, the rich history of human fire use in the SE USA by both the ancestors of modern Native Americans and Euro-American colonists alike complicates our understanding of the controls and drivers of fire in this region (Nowacki and Abrams 2008; Abrams and Nowacki 2015). Thus, there is significant potential benefit of paleofire-derived assessments of fire-climate and fire-vegetation relationships in this region.

Conclusions

We analyzed the morphological and morphometric characteristics of sedimentary charcoal preserved in a sediment core from Lake Matoaka in the SE USA. By comparing these data with known historical land-use changes, we (1) made inferences about the fuel sourcing of certain charcoal morphotypes, (2) identified the potential controls of several charcoal morphometric characteristics, and (3) proposed several modifications to an existing charcoal morphological key for application in the SE USA. We found that despite relatively consistent charcoal accumulation rates in the Matoaka sediments over the last 300 years, the morphology and morphometry of particles changed in response to broad-scale shifts of historical land use and population variations. Namely, we find that charcoal morphologies indicative of agricultural burning coincided with agricultural expansion and development prior to the American Revolutionary War ca. 1780 CE. Similarly, during the Antebellum Period preceding the American Civil War (1780 to 1865 CE), charcoal in Lake Matoaka reflects the documented reforestation which occurred in response to depopulation and economic depression. Following the Civil War (after ca. 1865), Williamsburg's population began to increase. This increase surged in the early 1900s, which was accompanied by increasing development which lasted until the modern. These increases of population and development were archived by changing charcoal morphologies. From the correspondence of these historical variations with morphotype shifts, we find evidence that Types M and S/B charcoal are likely sourced from burning herbaceous fuels. The variability of morphometric characteristics within classified morphotypes of charcoal preserved within the Lake Matoaka sediments suggests that morphotypes are unlikely to be sourced from sole fuel sources. Overall, we find that Type C charcoal may be too broadly defined for reliable use in the SE USA, and as such, we propose three additional morphotypes, Types AI (angular and irregular shaped particles), E (irregular, complex shaped particles), and T (triangular particles), which can help to disaggregate and better characterize shifting charcoal assemblages. More broadly, our analysis highlights that more work is needed to establish paleofire methodologies in the SE USA.

Acknowledgements We thank Elizabeth A. Canuel for her guidance and advice. We thank Dr. Thomas Whitmore, Dr. Jaime Escobar, and two anonymous reviewers for their helpful comments and improvements to this manuscript. This work was supported by honors fellowships granted to LFD and KC from the Charles Center at the College of William & Mary. LFD was also supported by a Stofan Scholarship from the Geology Department at William & Mary. RSV was supported by the Mellon postdoctoral fellowship in the Environmental Science and Policy Program at William & Mary. Support was also provided through National Science Foundation Grant EAR-1660309 to NLB and JMK.

References

Abrams MD, Nowacki GJ (2015) Exploring the early Anthropocene burning hypothesis and climate-fire anomalies for the eastern US. J Sustain for 34:30–48

Aleman JC, Blarquez O, Bentaleb I et al (2013) Tracking land-cover changes with sedimentary charcoal in the Afrotropics. Holocene 23:1853–1862

Balascio NL, Kaste JM, Meyer MG et al (2019) A high-resolution mill pond record from eastern Virginia (USA) reveals the impact of past landscape changes and regional pollution history. Anthropocene 25:100190

Blarquez O, Talbot J, Paillard J et al (2018) Late Holocene influence of societies on the fire regime in southern Québec temperate forests. Quat Sci Rev 180:63–74

Cahoon K (2019) Spheroidal carbonaceous particles in a Virginia mill pond provide a record of local and regional coal combustion

Cheung AH, Vachula RS, Clifton E et al (2021) Humans dominated biomass burning variations in Equatorial Asia over the past 200 years: evidence from a lake sediment charcoal record. Quat Sci Rev 253:106778

Clark JS (1988) Particle motion and the theory of charcoal analysis: source area, transport, deposition, and sampling. Quat Res 30:67–80. https://doi.org/10.1016/0033-5894(88)90088-9

Cohen JD (2000) Preventing disaster: home ignitability in the wildland–urban interface. J for 98:15–21

Constantine IV M, Mooney S (2021) Widely used charcoal analysis method in paleo studies involving NaOCl results in loss of charcoal formed below 400 C. Holocene 09596836211041740

Crawford AJ, Belcher CM (2014) Charcoal morphometry for paleoecological analysis: the effects of fuel type and transportation on morphological parameters. Appl Plant Sci 2:1400004

Cyrus C (2016) Floristic change spanning 45 years of global change in the College Woods, Williamsburg, Va

Daniau A-L, Sánchez-Goñi MF, Beaufort L et al (2007) Dansgaard-Oeschger climatic variability revealed by fire emissions in southwestern Iberia. Quat Sci Rev 26:1369–1383

Enache MD, Cumming BF (2006) Tracking recorded fires using charcoal morphology from the sedimentary sequence of Prosser Lake, British Columbia (Canada). Quat Res 65:282–292. https://doi.org/10.1016/j.yqres.2005.09.003

Evans MJ, Packard H (2007) Impact of urbanization on sediment chemistry in small-scale watersheds, southeast Virginia. In: AGU Fall Meeting Abstracts, pp H43D-1618

Feurdean A (2021) Experimental production of charcoal morphologies to discriminate fuel source and fire type in the Siberian taiga. Biogeosciences 18:3805–3821

Genet M, Daniau A-L, Mouillot F et al (2021) Modern relationships between microscopic charcoal in marine sediments and fire regimes on adjacent landmasses to refine the interpretation of marine paleofire records: an Iberian case study. Quat Sci Rev 270:107148

Handler R, Gable E (1997) The new history in an old museum. Duke University Press

Higuera PE, Whitlock C, Gage JA (2011) Linking tree-ring and sediment-charcoal records to reconstruct fire occurrence and area burned in subalpine forests of yellowstone National Park, USA. Holocene 21:327–341. https://doi.org/10.1177/0959683610374882

Hudspith VA, Belcher CM, Kelly R, Hu FS (2015) Charcoal reflectance reveals early Holocene boreal deciduous forests burned at high intensities. PLoS ONE 10:e0120835

Jaworski T (2017) World War II and the industrialization of the American South. J Econ Hist 77:1048–1082

Jensen K, Lynch EA, Calcote R, Hotchkiss SC (2007) Interpretation of charcoal morphotypes in sediments from Ferry Lake, Wisconsin, USA: do different plant fuel sources produce distinctive charcoal morphotypes? Holocene 17:907–915

Leys B, Brewer SC, McConaghy S et al (2015) Fire history reconstruction in grassland ecosystems: amount of charcoal reflects local area burned. Environ Res Lett 10:114009

Leys BA, Commerford JL, McLauchlan KK (2017) Reconstructing grassland fire history using sedimentary charcoal: Considering count, size and shape. PLoS ONE 12:e0176445

Leys BA, Marlon JR, Umbanhowar C, Vannière B (2018) Global fire history of grassland biomes. Ecol Evol 8:8831–8852

Marlon JR (2020) What the past can say about the present and future of fire. Quat Res 96:66–87

Marlon JR, Bartlein PJ, Carcaillet C et al (2008) Climate and human influences on global biomass burning over the past two millennia. Nat Geosci 1:697–702. https://doi.org/10.1038/ngeo313

Marlon JR, Bartlein PJ, Gavin DG et al (2012) Long-term perspective on wildfires in the western USA. Proc Natl Acad Sci USA 109:E535–E543. https://doi.org/10.1073/pnas.1112839109

Masiello CA (2004) New directions in black carbon organic geochemistry. Mar Chem 92:201–213. https://doi.org/10.1016/j.marchem.2004.06.043

Monette R, Ware S (1983) Early forest succession in the Virginia Coastal Plain. Bull Torrey Bot Club 110:80–86

Morgan TE (2004) Williamsburg: a city that history made. Arcadia Publishing

Mustaphi CJC, Pisaric MFJ (2014) A classification for macroscopic charcoal morphologies found in Holocene lacustrine sediments. Prog Phys Geog 38:734–754

Nowacki GJ, Abrams MD (2008) The demise of fire and "mesophication" of forests in the eastern United States. Bioscience 58:123–138

Pereboom EMB, Vachula RS, Huang Y, Russell J (2020) The morphology of experimentally produced charcoal distinguishes fuel types in the Arctic tundra. Holocene 30:1091–1096. https://doi.org/10.1177/0959683620908629

Power MJ, Marlon J, Ortiz N et al (2008) Changes in fire regimes since the Last Glacial Maximum: an assessment based on a global synthesis and analysis of charcoal data. Clim Dyn 30:887–907. https://doi.org/10.1007/s00382-007-0334-x

Radeloff VC, Hammer RB, Stewart SI et al (2005) The wildland–urban interface in the United States. Ecol Appl 15:799–805

Rehn E, Rehn A, Possemiers A (2019) Fossil charcoal particle identification and classification by two convolutional neural networks. Quat Sci Rev 226:106038

Rehn E, Rowe C, Ulm S et al (2021) Integrating charcoal morphology and stable carbon isotope analysis to identify non-grass elongate charcoal in tropical savannas. Veg Hist Archaeobot 31:37–48
Roos CI, Williamson GJ, Bowman DMJS (2019) Is anthropogenic pyrodiversity invisible in paleofire records? Fire 2:42
Rose NL (1994) A note on further refinements to a procedure for the extraction of carbonaceous fly-ash particles from sediments. J Paleolimnol 11:201–204
Schlachter KJ, Horn SP (2010) Sample preparation methods and replicability in macroscopic charcoal analysis. J Paleolimnol 44:701–708
Snitker G (2020) The Charcoal Quantification Tool (CharTool): a suite of open-source tools for quantifying charcoal fragments and sediment properties in archaeological and paleoecological analysis. Ethnobiol Lett 11:103–115
Stephenson MA (1947) Mills in eighteenth century Virginia: with special study of mills near Williamsburg
Terlizzi T (2021) Three centuries of vegetation change in the William & Mary College Woods reconstructed using Phytoliths
Theobald DM, Romme WH (2007) Expansion of the US wildland–urban interface. Landsc Urban Plan 83:340–354
Umbanhowar CE, McGrath MJ (1998) Experimental production and analysis of microscopic charcoal from wood, leaves and grasses. Holocene 8:341–346. https://doi.org/10.1191/095968398666496051
Vachula RS (2019) A usage-based size classification scheme for sedimentary charcoal. Holocene. https://doi.org/10.1177/0959683618816520
Vachula RS (2021) A meta-analytical approach to understanding the charcoal source area problem. Palaeogeogr Palaeoclimatol Palaeoecol 562:110111. https://doi.org/10.1016/j.palaeo.2020.110111
Vachula RS, Richter N (2018) Informing sedimentary charcoal-based fire reconstructions with a kinematic transport model. Holocene 28:173–178. https://doi.org/10.1177/0959683617715624
Vachula RS, Russell JM, Huang Y, Richter N (2018) Assessing the spatial fidelity of sedimentary charcoal size fractions as fire history proxies with a high-resolution sediment record and historical data. Palaeogeogr Palaeoclimatol Palaeoecol 508:166–175. https://doi.org/10.1016/j.palaeo.2018.07.032
Vachula RS, Russell JM, Huang Y (2019) Climate exceeded human management as the dominant control of fire at the regional scale in California's Sierra Nevada. Environ Res Lett. https://doi.org/10.1088/1748-9326/ab4669
Vachula RS, Sae-Lim J, Li R (2021) A critical appraisal of charcoal morphometry as a paleofire fuel type proxy. Quat Sci Rev 262:106979