

Sedimentary Accumulation of Black Carbon on the East Coast of The United States

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Abstract

The occurrence, trends and sources of soot black carbon (BC) in coastal sediments are poorly understood, particularly during the Anthropocene. Two sediment cores, covering the last ~ 100 years from the US East Coast, off North Carolina and in the Florida Straits, were analyzed for organic carbon (OC), BC fluxes and BC sources. BC fluxes were $0.1 \text{ g cm}^{-2} \text{ year}^{-1}$ at both sites and accounted for 8-22% of total OC. Carbon stable isotope values indicated OC to be of marine origin, while the BC was mostly terrestrially derived, C3-plant material. Radiocarbon values revealed BC originating mostly from fossil fuels or pre-aged carbon (fraction modern of 14–31%) at North Carolina, while in the Florida Strait the BC was mostly derived from biomass burning (fraction modern of 70–74%), in-line with continental (NC) or marine (FS) air mass origins. Ratios of polycyclic aromatic hydrocarbons broadly supported different BC sources at the two sites.

Key Points:

- Black Carbon fluxes were $0.1 \text{ g cm}^{-2} \text{ year}^{-1}$ at both Northeast shelf sites, accounting for 8-22% of total carbon.
- North Carolina had mostly old black carbon (^{14}C fraction modern 14-31%), likely from fossil fuel combustion.
- Black Carbon in the Florida Strait sediment was mostly biomass-derived (^{14}C fraction modern $\sim 70\%$), likely reflecting biomass burning

Plain Language Summary

Black carbon particles are the byproduct of incomplete combustion, either from wildfires or the deliberate burning of biomass and fossil fuels. Given its chemical and physical properties, black carbon particles are stable and accumulate in sediment. In coastal sediments, the black carbon composition will reflect changes in the burning of different fuels over time. This study provides information of how sources and intensity of black carbon deposition changed over a ~ 100 year period using sediment cores from two sites along the US East Coast. The result suggests black carbon deposition increased over time and was dominated by fossil fuel combustion around the North Carolina, while in Florida Strait the black carbon production did not change through period (1930s-2020s) and was influenced by vegetation burning.

1. Introduction

The Anthropocene indicates the era of increasing and dominating human activity on Earth (Crutzen, 2002), certainly noticeable in geological records since around the ~1950s, when emissions of black carbon (BC), and other pyrogenic tracers, such as polycyclic aromatic hydrocarbons (PAHs) started to rise, linked to increased industrial growth (Lima et al., 2003). BC in particular is among the most stable fractions of carbon and has received attention due to its widespread distribution in the environment, effects on climate change, and long residence time (Lund et al., 2018). The BC continuum includes a range of products, such as (in order of decreasing recalcitrance) soot, charcoal, char, and charred biomass (Masiello, 2004).

BC is emitted predominantly on land, as a byproduct of natural wildfires and the burning of fossil fuels and biomass (Masiello and Druffel, 2001), before it enters the ocean by atmospheric and riverine deposition (Jaffe et al., 2013; Mitra et al., 2002). In the Earth System, BC contributes around $12 \pm 5\%$ of the organic carbon (OC) in the rivers (Jones et al., 2020), 9-25% of OC in deltaic vegetated coastal habitat (Li et al., 2021), 3-15% in oceanic shelf sediment (Gustafsson and Gschwend, 1998), and 10-35% in deep ocean sediment (Lohmann et al., 2009). A recent focus on “dissolved black carbon” traces the presence of polycyclic aromatic moieties as part of the dissolved organic carbon cycle (Dittmar, 2008; Druffel et al., 2016), but whether this dissolved BC will reach and be stored in sediment remains unclear. To estimate BC burial fluxes and assess the influence of atmospheric and oceanic transport on the carbon cycle, sedimentary BC deposition and its sources ought to be investigated in the coastal area.

Currently very few measurements exist on fluxes of BC in the US coastal shelf (Gustafsson and Gschwend, 1998; Mitra et al., 2002; Yang and Guo, 2014). Typically, these studies have relied on a combination of stable C isotopes (as $\delta^{13}\text{C}$) with ratios of polycyclic aromatic

hydrocarbons (PAHs) to identify possible BC sources. Furthermore, differences in BC isolation methods (chemothermal oxidation at 375°C (CTO-375) vs. benzene polycarboxylic acid (BPCA) limit intercomparison between sedimentary BC studies (Hammes et al., 2007).

Only few studies have used the CTO-375 method and radiocarbon (as ^{14}C) to apportion sources of BC (biomass or fossil fuel) in aerosols along the East Coast of the US (e. g., Currie et al., 1997; Wozniak et al., 2012; Briggs and Long, 2016). These typically indicate the preponderance of fossil fuel burning in urban areas (Currie et al., 1997), and a roughly equal contribution of contemporary and fossil fuels in the aromatic fraction of aerosols (Wozniak et al., 2012).

Here, we present information from two sediment cores off the U.S. East Coast to determine changes in BC deposition and its sources over the last hundred years. Based on the above, we assumed that fossil fuel emissions were dominating sedimentary BC along the East Coast, but we wanted to test this in cores from contrasting sites – one representing the westerly trade winds, located off North Carolina, the other representing the northeasterly trade winds, located off Florida. This work was guided by the following objectives (1) to investigate distributions in sedimentary BC at two contrasting coastal sites, (2) to identify BC sources using a combination of $\delta^{13}\text{C}$ and ^{14}C values and PAH ratios, and (3) to determine sedimentary BC trends over time in the US East Coast.

2 Study Area and Methods

2.1 North Carolina Shelf and Florida Straits

Average wind (Figure S1) and Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) models (Figure S2) were compiled to determine air mass back origin. At the North Carolina (NC) core location, the wind was mostly coming from the south during spring-summer period, at least partially marine in origin, though mixing with coastal air masses can be expected. During the fall-winter period, air masses came from the north, reflecting US continental origin through atmospheric transport. On the other hand, at the Florida Straits core location, the air mass origin was mostly from the east and a small amount ($<8\%$) from the south side indicating both marine and terrestrial air mass origins.

Once in the water column, sediment transport in the North Atlantic is influenced by seasonal storms, wave-driven littoral currents, and the landward-oriented orbital wave current (Swift et al., 2009). During a nor-easter, the wind-driven component intensifies and is associated with downwelling. These flows can carry sediment to the shoreface if the waves are intense during the storm. Transport by storm currents is alongshore and offshore since wave orbital currents on the lower shoreface lack landward asymmetry.

2.2. Sample collection

Sediment samples were collected from the US East Coast at two sites: (1) Coastal North Carolina shelf (NC) and (2) in the Florida Straits (FS) during R/V Endeavor 597 cruise from May 1-7, 2017. The North Carolina Station was located between the Neuse and Cape Fear River basin, 90 km from Morehead City, while the FS core was located 45 km from Vero beach (see

Figure 1). Both the NC and FS coring sites were located on the continental shelf, at 200-400 meters depth.

At each site, sediment cores of approximately 15–30 cm were collected using a sediment multicore sampler. Each core was sectioned into 1 or 2 cm thick slices and preserved at -20°C . Prior to analysis, the sediments were placed in pre-combusted jars, dried in the oven at 60°C for 24 hours, and homogenized with mortar and pestle.

2.3 Black and Organic Carbon Concentration

The BC quantification followed the CTO-375 method to isolate soot BC in sediment (Gustafsson et al., 1997). Total organic carbon (TOC) was measured in duplicates by removing the inorganic carbon with acid treatment. Both soot BC and TOC were analyzed using a Costech 4010 Elemental Analyzer. The blank samples (pre-combusted sand) showed zero BC and TOC present. The SRM 1941b results were $6.1 \pm 0.63 \text{ mg g}^{-1}$ ($n=4$) for the BC and $30 \pm 0.46 \text{ mg g}^{-1}$ ($n=2$) for the TOC. Our measurement for SRM 1941b were similar to those reported in the literature: $5.8 \pm 0.5 \text{ mg g}^{-1}$ for BC (Gustafsson et al., 2001), $5.2 \pm 1.4 \text{ mg g}^{-1}$ for BC (Lohmann et al., 2009), $5.7 \pm 0.28 \text{ mg g}^{-1}$ for BC (Elmqvist et al., 2006), and $30 \pm 2.4 \text{ mg g}^{-1}$ for TOC (Gonzalez and Watters, 2015).

2.4 Lead-210

One of the important factors to be able to calculate BC fluxes is deriving the sedimentation rates, which were determined using ^{210}Pb age models. We used a Canberra Ultra-Low Background Gamma Spectrometer (model GCW3023) to measure the ^{210}Pb , ^{226}Ra , and

^{137}Cs activity from dry sediment samples, with Analytics Mixed Gamma SRD51276-399 standard used for instrument calibration. The calibration check for the sample calculation was performed using NIST SRM 4357 and IAEA 315. The sedimentation rate was calculated based on unsupported /excess ^{210}Pb in each sediment layer as it declined exponentially with depth. Sedimentation rates were estimated from the constant initial concentration (CIC) linear accumulation model, which assumed a steady input of the sediment over time (Appleby, 1993).

2.5 Stable Carbon Isotope ($\delta^{13}\text{C}$) Analysis for BC and TOC

Stable carbon isotopes were used to assess compositional differences between BC and TOC sources. $\delta^{13}\text{C}$ values were measured on an Elemental Analyzer - Isotope Ratio Mass Spectrometer (EA-IRMS; Costech 4010 EA coupled with a Thermo Delta V Advantage). $\delta^{13}\text{C}$ values are reported in parts per mil (‰) relative to the reference standard Vienna Pee Dee Belemnite (VPDB) (Ceburnis et al., 2011). An in-house aminocaproic acid standard was used for the linearity correction during sample analysis. The standards for instrument offset correction included USGS 65 and Urea from IVA Analysen Technik for C isotopes. $\delta^{13}\text{C}$ values had standard deviation better than $\pm 0.4\text{‰}$ based on repeated measurements of duplicate samples.

2.6 Radiocarbon Analysis ($\Delta^{14}\text{C}$) for BC

$\Delta^{14}\text{C}$ values were used to distinguish the age and fossil fuel / non-fossil fuel contribution of BC. Measurements, performed by the National Ocean Sciences Accelerator Mass spectrometry (NOSAMS) facility, were conducted on BC sample. Radiocarbon was reported as

‰ deviation ($\Delta^{14}\text{C}$) relative to NBS Oxalic Acid I (NIST-SRM-4990) (Stuiver and Polach, 1977).

2.7 PAH Analysis

PAHs measurements were used to identify the origin of soot BC due to the virtue of changing compound ratios as a function of combustion sources, following a modified EPA method and QA/QC (GCMS analysis; protocols: 8270D). The PAHs were quantified using a GCMS Agilent 6890 Series GC System coupled with Agilent 5973 Network Mass Selective Detector. Mean recoveries of the internal standards were 20% for d_{10} -Acenaphthene, 29% for d_{10} -Phenanthrene, and 50% for d_{12} -Chrysene. PAH concentrations in laboratory blanks were below detection limits.

3 Results

3.1 Black and Organic Carbon

We isolated soot BC using the CTO-375 method to obtain the combustion-derived deposition history of soot BC. This method provides information indicative of long-range (atmospheric) transport because of the soot size as opposed to charcoal, char, and charred biomass (Pohl et al., 2014). In general, the BC concentrations were $< 1 \text{ mg g}^{-1}$ at both sites (NC: $0.3\text{--}0.7 \text{ mg g}^{-1}$ and FS: $0.5\text{--}0.8 \text{ mg g}^{-1}$) (see Figure 2). At NC, BC displayed two peaks in the recent past, one from 2012 - 2017, and another spike in 2004 (0-2 cm and 3.5 cm with $0.6\text{--}0.7 \text{ mg g}^{-1}$ BC). At FS, BC concentrations were mostly stable over time; greater concentrations of BC with 0.8 mg g^{-1} BC were present in the late 1930s, early 1990s, and late 2010s (26.5cm, 8.5

cm, and 0.5 cm). Our BC concentrations were lower than reported for the Pettaquamscutt River basin ($\sim 4.7\text{--}6.1\text{ mg g}^{-1}$) (Hanke et al., 2016), however within range of BC value reported for North Atlantic shelf ($\sim 0.11\text{--}1.7\text{ mg g}^{-1}$) (Gustafsson and Gschwend, 1998), for the Palos Verdes shelf ($\sim 0.5\text{ to }1.6\text{ mg g}^{-1}$) (Middelburg et al., 1999), and in deep South Atlantic sediments ($\sim 0.4\text{--}1.7\text{ mg g}^{-1}$) (Lohmann et al., 2009).

The TOC concentrations in the NC core were lower than in the FS, with $3.1\text{--}4.7\text{ mg g}^{-1}$ and $6.3\text{--}7.8\text{ mg g}^{-1}$, respectively. Both profiles displayed peaks in the recent past – one in the late 1980s, and more recently in the early and late 2010s: The highest peak at NC was found in 1988 (4.7 mg g^{-1}), while the highest peak at FS was found in 1995 (7.8 mg g^{-1}). Moreover, our results were almost similar to TOC in the South Atlantic Ocean (Lohmann et al., 2009) whereas it is lower compared with the Gulf of Maine (Gustafsson and Gschwend, 1998).

BC/TOC ratios (see Figure S4 in Supporting Information S1) were higher at NC, with increasing concentrations from 1930s to 2020s (9-22% for NC and 8-12% for FS), while BC/TOC ratios remained roughly constant at FS. The pattern suggests more refractory carbon in the form of BC was present at NC. The results were similar to those from deltaic vegetated coastal habitat (9-25%) (Li et al., 2021), and within the range reported for global rivers ($12 \pm$

5%) (Jones et al., 2020) and oceanic shelf sediment (3-15%) (Gustafsson and Gschwend, 1998), but lower than deep ocean sediment (10-35%) (Lohmann et al., 2009).

3.2 Sedimentation rates

The average sedimentation rates for NC and FS were 0.28 cm yr^{-1} and 0.33 cm yr^{-1} , respectively (see Figure S3 Supplementary Information), within the range observed for other East Coast shelf sites. For example, reported sedimentation rates at the continental shelf of Cape Hatteras ranged from ~ 0.01 to 0.43 cm yr^{-1} (Alperin et al., 2002), from ~ 0.03 to 0.15 cm yr^{-1} for the continental slope of Cape Hatteras (Thomas et al., 2002), and ~ 0.1 and 0.16 cm yr^{-1} for the upper slope western Great Bahama Bank (Toomey et al., 2013). No ^{137}Cs was present in our samples. This is not unusual, as it was also not observed in other locations along the east coast of the US (Lima et al., 2003). The absence of ^{137}Cs could also be due to the integration of $>2 \text{ cm}$ depth intervals for this work.

3.3 BC Accumulation Rates/Fluxes

BC accumulation rates ($\mu\text{g cm}^{-2} \text{ year}^{-1}$) were calculated by multiplying dry bulk density, estimated from a generic value for US East coastal sediment ($\sim 0.9 \text{ g cm}^{-3}$) (Boggess et al., 2021), with BC concentration determined here ($0.3\text{-}0.7 \text{ mg g}^{-1}$ for NC and $0.5\text{-}0.8 \text{ mg g}^{-1}$, Figure 2), and average sedimentation rates from the $^{210}\text{Pb}_{\text{xs}}$ profiles ($0.28 \text{ cm year}^{-1}$ for NC and $0.33 \text{ cm year}^{-1}$ for FS, see Figure S3 in Supporting Information S1). Based on these calculations, BC fluxes varied from $87\text{-}170 \mu\text{g cm}^{-2} \text{ year}^{-1}$ for NC, and from $150\text{-}250 \mu\text{g cm}^{-2} \text{ year}^{-1}$ for FS. The

BC distribution at both sites showed different patterns - the BC was higher at the surface in NC, while there was little variation with depth at FS. The accumulation rates of BC were lower than South Atlantic BC sediment fluxes ($500 - 780 \mu\text{g cm}^{-2} \text{ year}^{-1}$) (Lohmann et al., 2009), but within range of North Atlantic sediments (~ 90 to $190 \mu\text{g cm}^{-2} \text{ year}^{-1}$) (Gustafsson and Gschwend, 1998).

3.4 Stable Isotope $\delta^{13}\text{C}$ ratio

The $\delta^{13}\text{C}$ values of the BC fractions were more depleted in NC than FS (Figure 2). Sedimentary BC $\delta^{13}\text{C}$ values ranged from -21.3 to -23‰ in NC, and -20.2 to -21.9‰ in FS, while TOC $\delta^{13}\text{C}$ values were around -19.8 to -20.9‰ in NC and -19.5 to -20.3‰ in FS, respectively. Importantly, there was a constant offset of $\sim 2 \text{‰}$ between BC and TOC for both cores, indicating that BC and TOC represented two different carbon pools. Based on these $\delta^{13}\text{C}$ values, we assign terrestrial (C3) sources to the NC BC fraction ($\delta^{13}\text{C} > -22\text{‰}$), while a combination of C3 and oil derived from plankton ($\delta^{13}\text{C} < -22 \text{‰}$) dominated at FS (Bush et al., 2007; Holtvoeth et al., 2003). According to our results, C4 plant emissions did not contribute substantially to the BC formation.

3.5 Radiocarbon $\Delta^{14}\text{C}$

$\Delta^{14}\text{C}$ values provide the best evidence to distinguish between modern biofuels or young carbon ($+62\text{‰}$) and fossil fuel or old carbon (-1000‰) (Ruppel et al., 2021; Wozniak et al., 2012). $\Delta^{14}\text{C}$ values ranged from -697 to -863‰ (fraction modern of 14-31%), indicate the predominance of fossil fuels at NC. In the FS core, $\Delta^{14}\text{C}$ values ranged from -268 to -307‰ (fraction modern of 70-74%), implying the BC is mostly composed of biofuels or young carbon.

Interestingly, these radiocarbon results are the strongest indication, among all the tracers used in this study, that the two cores had a different BC origin.

3.6 PAHs

PAH concentrations reflected a general increase in production and consumption of fossil fuel, including atmospheric transport as PAHs are emitted from upwind source regions and maritime (Hanke et al., 2016; Lohmann et al., 2009a). Total PAH concentrations (sum of phenanthrene, anthracene, flouranthene, pyrene, benzo(*a*)anthracene, chrysene, benzo(*b*)flouranthene, benzo(*k*)fluoranthene, benzo(*e*)pyrene, and benzo(*a*)pyrene) were lower in the NC core, ranging from 0.08 ng g⁻¹ dw (mid 1930s and early 2010s) to 1.38 ng g⁻¹ dw (mid 1990s), than in the FS, ranging from 0.43 ng g⁻¹ dw (early 1950s) to 1.14 ng g⁻¹ dw (mid 1990s), except in one interval in the mid 1990s where TPAH reached similar peaks (NC around 1.38 ng g⁻¹, while FS with 1.14 ng g⁻¹). The Organization of Petroleum Exporting Countries (OPEC) oil embargo in 1973 might have contributed to slight decreases in TPAH in FS from the 1970s to 1980s (Lima et al., 2003). The same peak in mid 1990s in both locations presumable reflected a larger rise in diesel consumption between 1996 and 1999, as reported Lima et al., (2003). TPAHs decreased steadily after mid 1990s until reach the lowest concentration, similar to the 1950s period, before increasing again in the late 2010s.

The analysis of PAHs isomers ratios (see Figure 3) can also be used to gain more information on the origin of soot BC via the associated PAH diagnostic ratios (Yunker et al., 2002, 2011). Characteristic PAH ratios indicated the predominance of biomass and petroleum combustion sources. Most samples at both sites indicated a mixture of biomass and petroleum combustion, with one sample from Florida reflecting petroleum dominance. The coal industry

was dominant in the nineteenth century, before petroleum products overtook it, as natural gas usage increased rapidly (Lima et al., 2003).

4 Discussion

BC accumulation off the US East Coast was evaluated in two steps. First, the temporal variation of BC and TOC accumulation were measured at two sites, and fluxes derived. Second, carbon isotope and PAH ratios were assessed to identify sources of sedimentary BC were assessed over time.

4.1 Presence of Black and Organic Carbon

A study from Lima et al., (2003) reported increasing combustion activity (via concentrations of PAHs) from the 1950s, when the petroleum became the dominant energy sources, up to 1973 before the Organization of Petroleum Exporting Countries (OPEC) oil embargo began. Our result also indicated the BC emission after 2000 increased ten to twenty percent off US East coast compared with 1950s period. The results showed elevated BC/TOC ratio in NC coincided with lower TOC concentrations which may be attributed to elevated BC supply retained in sandy sediment with low organic carbon content at NC (Chew and Gallagher,

2018). According to Gustafsson and Gschwend (1998), BC fluxes on the continental shelves could account for 40% of global BC emissions.

BC accumulation rates also reflect the sequestration capacity of the shelf sediment. Results presented here imply the sequestration capacity of shelf sediments should also take into account allochthonous BC/refractory carbon, not only the autochthonous (organic) carbon.

4.2 Origin of Black Carbon

Based on the location, the US East Coast might receive BC input from maritime fossil fuel emissions, and atmospheric transport from the continental US and the Bahamas. Ocean currents along the US East Coast are probably not affected by oil production activity (and emission) around the Gulf of Mexico. Due to the surface prevailing easterly winds, the drift pushes any oil releases around the Gulf of Mexico to reach the coast along the northern Gulf, and the Loop Current evolution prevented oil from reaching the Florida Strait (le Hénaff et al., 2012). So, most likely, oil pollution from the Gulf of Mexico will not reach the East Coast, and the absence of oil was confirmed by the ratios of PAHs, and their overall low concentrations in the sediment.

Continental and maritime sources contributed BC to the sediment cores, especially NC, which was enriched in $\delta^{13}\text{C}$ ratio representing C_3 plant and fossil fuels (Bush et al., 2007), while the OC fraction indicated a marine origin. Meanwhile, the average BC $\delta^{13}\text{C}$ values at FS sites seemingly emphasized marine sources but could reflect a combination of C_3 plant (Fall et al., 2021) and fossil derived (i.e., oil) marine sources (Wozniak et al., 2012). Wagner et al. (2019) suggested that the $\delta^{13}\text{C}$ signatures can undergo changes during photolytic exposures and other processes including sorption/ desorption during delivery to the bottom sediment, though

particulate BC is considered to be recalcitrant. Older $\Delta^{14}\text{C}$ ages at NC (9,520 – 15,900 years BP) and distinctive BC $\delta^{13}\text{C}$ compared to TOC $\delta^{13}\text{C}$ values also provided strong evidence that BC was produced from the burning of older biomass, predominantly C3 plant and delivered into the NC coast through atmospheric transport from the continent. According to a study of BC concentrations in the tropical Atlantic Ocean marine boundary layer, soot-aerosol is a significant contributor to deep ocean sediment (Pohl et al., 2014).

Based on the PAHs diagnostic ratio, each stations claimed a distinct general pattern. Temporal trends of PAH ratios at NC indicate a shift from biomass burning in the 1950s towards petroleum in the most recent sediment (~ 2017), in-line with expectations of an increasing use of liquid fossil fuels including gasoline and diesel after the second world war. This is somewhat reflected in the increasing fraction modern from ^{14}C results (fraction modern increasing from 14% in the most recent sediment to 31% in the deeper sediment layers). For the sediment layers at FS, on the other hand, PAH ratios indicate no major changes in biomass-burning over time (which is mirrored in the ^{14}C fraction modern results that vary from 70%-74% over time). Sedimentary accumulation fluxes of BC at FS did not display strong temporal trends downcore, in-line with a predominance of not-fossil fuel derived BC.

Based on latitude and air mass back trajectories, westerly winds affected the NC site, hence continental fossil fuel emissions can reasonably explain the predominantly fossil fuel origin of the BC. In contrast, the trade winds affected the FS site which can explain the different atmospheric BC transport and deposition patterns between both sites. At FS, the deposition

implies biomass burning, likely from the burning of biomass and forest/shrub wildfires from the Caribbean region, and possibly from Eastern Africa (Marufu et al., 2000).

If we combine the information from the $\delta^{13}\text{C}$, $\Delta^{14}\text{C}$ and PAHs diagnostic ratios, there is some congruence evident in the tracers. PAH ratios indicate the prevalence of petroleum combustion at NC, which is backed up by ^{14}C results (mostly fossil fuels), while $\delta^{13}\text{C}$ ratios implied mostly terrestrial C_3 plant emissions at NC. In contrast, at FS $\delta^{13}\text{C}$ values mostly indicated a marine signature, likely from the burning of petroleum, while PAH ratios implied a mixture of biomass and petroleum emissions, and ^{14}C results indicate the prevalence of biomass-derived BC.

5 Conclusions

In this study, we evaluated temporal trends of soot BC accumulation in two continental shelf sediment cores from the US East Coast. Trends of BC concentrations displayed two peaks in the last three decades, probably reflecting changes in traffic emissions. Stable carbon isotope ratios confirmed the marine origin of the OC fraction in the sediment, while BC was more depleted, and likely dominated by emissions from terrestrial C_3 plants and fossil fuels (Bush et al., 2007; Holtvoeth et al., 2003). PAH ratios indicate the prevalence of petroleum combustion at NC, which is backed up by ^{14}C results (mostly fossil fuels), while $\delta^{13}\text{C}$ ratios implied mostly terrestrial C_3 plant emissions at NC. In contrast, at FL $\delta^{13}\text{C}$ values mostly indicated a marine signature, likely from the burning of petroleum, while PAH ratios implied a mixture of biomass and petroleum emissions, and ^{14}C results indicate the prevalence of biomass-derived BC. Our

results support shows the importance of relying on multiple markers to constrain fluxes and sources of BC deposition in the shelf sediments during the Anthropocene.

CRedit authorship contribution statement

Ita Wulandari: Writing – original draft, Writing – review & editing, Visualization, Investigation. **Samuel Katz:** Methodology, Validation, Writing – review & editing. **Roger Patrick Kelly:** Writing – review & editing, Data curation, Formal analysis. **Rebecca Robinson:** Writing – review & editing. **Rainer Lohmann:** Conceptualization, Writing – review & editing, Funding acquisition.

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Open Research

The concentrations of organic, black carbon, polycyclic aromatic hydrocarbons and the isotope ratios of ^{210}Pb , ^{13}C , and ^{14}C used for the interpretation of Black carbon fluxes, trends and source apportionment in the study are available at Open Science Framework via:

<https://doi.org/10.17605/OSF.IO/DBSEU>

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Figures

Figure 1. Sediment core locations off North Carolina (NC) and in the Florida Straits (FS)

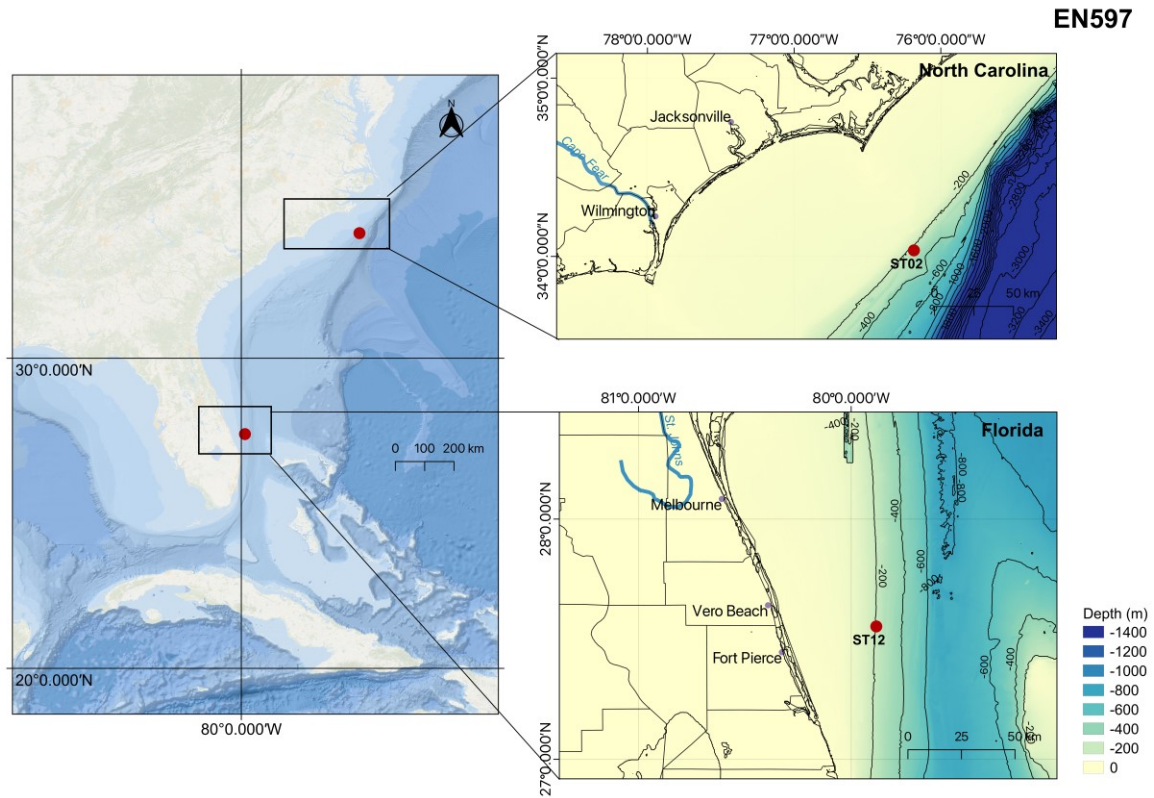


Figure 2. Sediment concentrations of black carbon (BC), total organic carbon (TOC), organic carbon (OC), and total polycyclic aromatic hydrocarbon (TPAH) (a, b, c, d) and $\delta^{13}\text{C}$ (e, f, g) and $\Delta^{14}\text{C}$ values (h) in North Carolina (NC) and Florida Straits (FS) shelf cores.

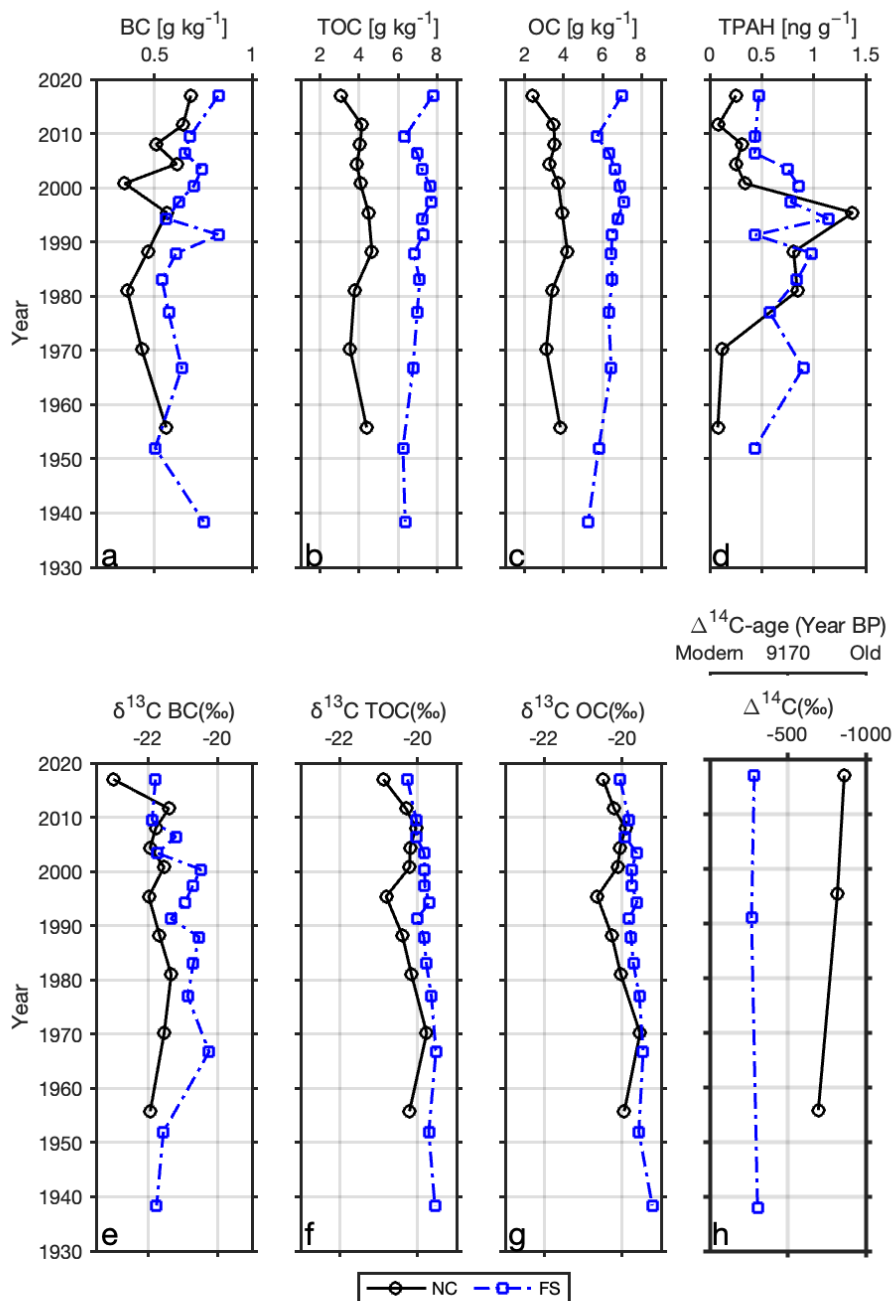


Figure 3. Cross plots of PAH ratios of (1) Fla/(Fla+Pyr) vs. Ant/(Ant+Phe), (2) Fla/(Fla+Pyr) vs. BaA/228, and (3) Fla/(Fla+Pyr) vs. Ant/178.

