

HISTORICAL ATMOSPHERIC LEAD AND OTHER TRACE
ELEMENTS DEPOSITION RECORDS IN AN OMBROTROPHIC PEAT
POCOSIN: A NEW RECORD FROM NORTH CAROLINA

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ABSTRACT

**JINGYUAN SUN: Historical Atmospheric Lead And Other Trace Elements
Deposition Records In An Ombrotrophic Peat Pocosin: A New Record From North
Carolina**

(Under the direction of Larry Benninger)

Human activities introduce pollutant elements into the atmosphere, from which they enter sedimentary environments via precipitation and dry deposition. In this study, pocosin peat in eastern North Carolina was used to track the sources of atmospherically deposited trace elements. Peat chronology was based on ^{210}Pb and fallout ^{137}Cs . Concentrations, accumulation rates and enrichment factors for twenty-five elements, along with Sr and Pb isotopic compositions, were obtained. The results show that local leaded gasoline usage dominated the lead source. However, other anthropogenic sources such as coal combustion, mining, and agricultural development may have also introduced Pb and other trace elements through windblown dust. Sr isotopes require sources more radiogenic than seawater Sr; twentieth century Pb isotopes are consistent with predominant input from gasoline lead and coal combustion. The pocosin record of lead is similar to published records from the eastern United States and Western Europe, but contrasts with those from Asia.

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Introduction

In the last several centuries, increasing industrial activity has introduced pollutant trace elements into the atmosphere on a global scale. These trace elements enter sedimentary environments via precipitation and dry deposition. The record of trace element deposition from the atmosphere can be tracked from ice cores (Candelone et al., 1994; Barbante et al., 2004), peat deposits (Shotyk, 1995; Coggins et al., 2006; Cloy et al., 2008) and lake sediments (Koinig et al., 2003). Due to the isolation of the peat deposition from nearby groundwater and surface water in most ombrotrophic bogs (Damman, 1986, 1987), the only water input to ombrotrophic peat bogs is precipitation; therefore the inorganic component of ombrotrophic peats is introduced from the atmosphere (Shotyk, 1995).

Peats are formed when decomposition rates are lower than rates of dead vegetation accumulation, and are commonly found in such places as swamps and marshes. A swamp is a wooded area where the soil is usually saturated or covered by surface water for a few months of the growing season; peaty swamps include bays, bogs and pocosins (Penfound, 1952). The word “pocosin” refers to freshwater, ever-green shrub bogs, and comes from an Algonquin Indian word which means “swamp-on-a-hill” (Tooker, 1899; Richardson, 1983). Because the elevation of pocosins is higher than that of the surrounding lakes and no divided drainage and streams are in the area, water from precipitation is expected to flow radially away from the pocosin (Phillips, 1997; Richardson, 2003).

In the last 20 years, long and short-term trace element records in European peatland studies suggest that estimated trace element concentrations, flux and accumulation rates not only relate to the historical anthropogenic major events in different times such as the Industrial Revolution, but also act as indicators of local industrial activities on shorter time scales. Such activities involve, for example, leaded gasoline usage and industrial mining activities (Shotyk et al., 1998; Vleeschouwer et al., 2007; Cloy et al., 2008). Therefore, similar peatland records from other areas such as North America and Asia may show quite different trace elements deposition profiles compared with the European studies within the same time periods (Kamenov et al., 2009; Bao et al., 2010).

In North Carolina, the regional anthropogenic activities related to trace elements emissions include leaded gasoline usage, mining, oil/gas exploration, coal combustion, forest fires, and agriculture over the last 100 years. Leaded gasoline was introduced in 1923 in the United States (Kamenov et al., 2009), while its phasing out began in 1973 (US EPA). As for the other activities, North Carolina has its own characteristics due to local geologic resources, regional population change and industrial development. The coastal plain of North Carolina (the focus of this study) is dominated by non-metal mineral mines which have extracted peat (for soil/fertilizer), sand and gravel, phosphate and crushed stone (limestone, granite, etc.) (Stuckey, 1965). However, coal mining (for 1918-1930, the greatest production was in the Cumnock and Carolina mines), heavy metal mining and gas and oil exploration (for 1925-1963, a total of 43 boreholes were drilled in the outer coastal plain) appeared for a short period but have never been important in the area (Stuckey, 1965; USGS). Furthermore, natural and anthropogenic

forest fires have been significant around the coastal forest for more than a century (Fowler and Konopik, 2007).

Major events in North Carolina are summarized in Table 1. Sand and gravel, crushed stone, and phosphate, as well as widespread agriculture, have all occurred along the east coast of North Carolina. Their byproducts are the major potential sources of mineral dust deposition in coastal pocosins. The North Carolina Geological Survey (NCGS) reports that the primary source of crushed stone in the coastal plain is limestone. In the same area, phosphate mines have been developed for about 60 years (USGS) (Figure 1). The historical populations and growth rates for the five major relevant counties are also shown in Figure 2 and 3.

Trace elements have been used as markers for different anthropogenic activities (Huang et al., 1994). Many studies have utilized lead (Pb) concentration profiles and accumulation rates to track historical local industrial activities such as leaded gasoline usage (Shotyk et al., 1998; Dunlap et al., 1999; Vleeschouwer et al., 2007). The spatial variability of these data imply regional lead pollution conditions (Rothwell et al., 2007; Novak et al., 2008). Also, lead isotope composition ($^{206}\text{Pb}/^{207}\text{Pb}$) has been compared among several areas as a signal to track other local industrial histories such as the sources of ore at different times (Shotyk et al., 1998; Bollhöfer and Rosman, 2001; Cloy et al., 2008; Kamenov et al., 2009).

Besides Pb, other trace elements (Cu, Zn, Cd, Sn, Sb, Bi, V, Ni) are also discussed in this study to develop a better understanding of the local history of anthropogenic activities. Numerous studies from the other regions have used trace elements as an

indicator of atmospheric pollutant deposition in peatlands (Cochran et al., 1998; Mighall et al., 2009; Kamenov et al., 2009; Olid et al., 2010). Major elements such as Al, Fe, Mg, Mn, and Ti and minor trace elements of low geochemical mobility (REE, Th) were also analyzed. These elements can trace inputs of dust from silicate and oxide sources, while the elements of low solubility in normal conditions are concentrated in the ash fraction of the peat (Weiss et al., 2009; Shotyk et al., 2002; Kamenov et al., 2009). Therefore, ash content is an important factor in interpreting the concentrations of some elements in peat. “Ash” refers to the inorganic materials of peat, and is made up of minerals and ionically bound inorganics (Andrejko et al., 1983). Elements deposit in peat in the form of ash via precipitation and dry deposition. Typical ash content for ombrotrophic peat is below 5%, whereas minerotrophic (river-fed) peat usually has ash contents well above 5% (Weiss et al., 1999).

To study the last 100-200 years of peat deposition, ^{210}Pb is commonly used to establish peat chronology (Appleby et al., 1997; Farmer et al., 2006; Novak et al., 2008; Cloy et al., 2008; Bao et al., 2010). Nuclear fallout ^{137}Cs has also been used with ^{210}Pb to help establish a more secure chronology (Oldfield et al., 1979; Oldfield et al., 1995; Appleby et al., 1997). ^{210}Pb and ^{137}Cs inventories in these studies reflect the regional ^{210}Pb and ^{137}Cs deposition (Oldfield et al., 1979).

This study adds a new record to North America peat studies with cores obtained from a pocosin in the Croatan National Forest (CNF), located in the coastal plain of North Carolina. Using peat chronology, an historical record of the deposition of major and trace elements is produced. Isotopic compositions of Pb and Sr are used to investigate aerosol sources to the pocosin. Comparison with previous studies suggests that during the 20th

century, the atmospheric source of Pb was likely dominated by aerosols produced from leaded gasoline. One goal of the study is to test whether North Carolina pocosin peats are well-suited for distinguishing the atmospheric sources of trace elements. The other goal is to test whether deposition in North Carolina during this time period is broadly similar to that observed in elsewhere in North America and in Western Europe. Different histories of usage of leaded fuels apply in Asia, and consequently different historical patterns in lead deposition may be expected. Differences in Pb deposition among North American and Western European sites should reflect differences in local and regional industrial activities.

Site description

On 12 March 2011, two peat cores were taken from an ombrotrophic pocosin in Croatan National Forest (CNF) (34°53'14"N, 077°06'30"W) (Figure 4). Near this study site, minor gravel roads and ditches have been built during the development of the forest area. The surface elevation of the peat horizon ranges from 9 to 12 meters above mean sea level, and the peat thickness ranges from 0 to 2.5 meters (Ingram, 1987). The pocosin is generally dominated by shrubs and some pine trees, and most common species of shrubs in CNF are *Arundinaria gigantea*, *Persea borbonia*, *Cyrilla racemiflora*, and *Ilex coriacea* (Richardson, 2003). The average annual precipitation at the closest town, Maysville (about 15 km west), is 142 cm. The annual average temperature ranges from -0.5 to 31°C (SERCC). Part of the CNF area is cut by ditches along Forest Service roads (Figure 4).

Methods

Field sampling

A 1.5 m long soil probe was used to test the thickness of the peat deposit in the study area. The sampling site was chosen when the testing probe easily reached deeper than one meter. Two peat cores were collected using stainless steel core tubes with inner diameters of 9.5 and 9.8 cm and a length of 50 cm. The two cores were located about 1.5 meters apart at the field site; Core 2 was at higher elevation by about 10-15 cm. Both cores were sealed immediately with rubber stoppers at both top and bottom. They were returned to Chapel Hill within six hours of collection and stored in a walk-in refrigerator (4°C) until subsampling.

Sample preparation

The loose leaves on the top of the cores were removed and bagged before subsampling the peat. The surface of Core 1 showed appreciable relief (about 3.5 cm). To create a flat surface the first subsample was therefore taken to level the core surface and assigned an average thickness of 1.75 cm. The second and third subsamples of Core 1 were about 1.4 cm thick. Core 1 was then sampled in 1-cm increments. The surface of Core 2 was relatively flat; subsamples 1 – 4 were 1.25 cm thick, and the remainder of the core was sampled in 1-cm increments. To avoid effects of mixing along the inner surface of core tube, the outer edges of each sample were trimmed off beginning at 18 cm depth

in Core 1 and 3.75cm depth in Core 2 (the beginning of trimming depth was based on the dryness and shape of the edge of each section). Leaves and roots were commonly found in the layers closest to the surface, and thicker roots were found at intervals through the entire cores. Peat depths containing roots were noted during sampling, and the larger roots were discarded. All samples were placed in pre-weighed polypropylene jars which were weighed and stored in a laboratory freezer (-12°C) after sampling in preparation for freeze drying. All the peat samples were freeze-dried at -40°C or below for 72 hours.

The water content of the peat was determined by re-weighing samples after freeze-drying. Bulk density (g cm^{-3}) was calculated from total dry mass of each sample divided by each increment volume. Increment volume was calculated from increment thickness and radius, after correcting the radius for any trimming (in Core 1, 1 mm for depth 18-22 cm, and 2 mm for depth > 22 cm). Ash content was determined by roasting dry peat samples in air. The furnace temperature was increased incrementally from 200 °C to 500 °C, and held over night at 500°C. Ash weights were recorded and calculated as loss on ignition, which is interpreted as organic matter content.

¹³⁷Cs and ²¹⁰Pb dating

¹³⁷Cs and ²¹⁰Pb activities were determined following the procedures described in Benninger and Wells (1993). All the dried samples were ground in a porcelain mortar and packed into polypropylene jars for gamma counting. Most samples (70 of 77) were counted in 1-oz polypropylene jars, while the rest were counted in 0.5-oz jars. The sealed samples were stored for more than two weeks before gamma counting on one of two intrinsic Ge detectors in the Department of Marine Sciences at UNC-CH. Detector

efficiencies were calibrated with standards prepared from by mixing certified reference materials (uraninite from EPA, monazite from New Brunswick Lab, ^{137}Cs from Amersham International, now GE Healthcare) into purified cellulose fibers. The peaks in gamma spectra were manually integrated above baselines and corrected for backgrounds averaged at bi-weekly intervals. ^{226}Ra was determined by its daughters ^{214}Pb at 295.1 keV and 352 keV and ^{214}Bi at 609.3 keV and ^{226}Ra presents *in situ* supported ^{210}Pb activity; ^{228}Ra was determined by its daughters ^{228}Ac at 338.4 keV and 911.2 keV; ^{228}Th was determined by ^{212}Pb at 238.6 keV and ^{208}Tl at 583 keV. ^{137}Cs was measured by its emission at 661.7 keV. Excess ^{210}Pb and ^{137}Cs were decay-corrected to the date of collection using half-lives of 22.3 years and 30.1 years, respectively. The excess/unsupported ^{210}Pb is obtained from total ^{210}Pb (^{210}Po activity) minus supported ^{210}Pb (^{226}Ra activity).

Total ^{210}Pb activity was determined via its granddaughter ^{210}Po on alpha spectrometers in the Department of Geological Sciences. The methods of laboratory preparation and digestion were similar to the work of Benninger and Wells (1993). Five to 10 gr samples of dry peat were spiked with ^{209}Po , and went through complete acid digestion in 10 mL concentrated HNO_3 , 2 mL HF, and 4 mL 1:1 sulfuric acid/water acid on a hotplate for 4 days on average. An additional 5 mL concentrated HNO_3 and 20 drops of 30% H_2O_2 were added 1-3 times during digestion to promote complete dissolution. The solution was evaporated to dryness and dissolved by heating into 50 mL 0.2N HNO_3 . Any residual particulate matter was removed by filtering through Whatman 44 paper. Iron carrier was added to the filtrate and polonium isotopes were scavenged by

precipitation of hydroxides at pH 7-8. After dissolution of the precipitate into dilute HCl, Po was deposited onto silver disks for alpha spectrometry.

The CRS model for ^{210}Pb dating was applied in this study and ^{137}Cs was used as a single chronology marker within the core (Appleby and Oldfield, 1978; Oldfield et al., 1979; Robbins and Herche, 1993; Cochran et al., 1998). The constant rate supply of ^{210}Pb (CRS) model is most suitable to produce this chronology; the underlying assumptions are that unsupported ^{210}Pb flux is constant and that no obvious migration of ^{210}Pb is observed in the sediment (Appleby and Oldfield, 1978; Robbins, 1978). Using CRS (Robbins and Herche, 1993), the age t (y) is obtained by:

$$t = - (1/\lambda) \ln [1 - \Sigma^{210}\text{Pb}(z)/\Sigma^{210}\text{Pb}(\infty)] \quad (1)$$

where $\Sigma^{210}\text{Pb}(\infty)$ is the total excess ^{210}Pb inventory (Bq m^{-2}), $\Sigma^{210}\text{Pb}(z)$ is the accumulative excess ^{210}Pb activity (Bq m^{-2}) at the depth z (cm). λ is decay constant of ^{210}Pb ($\lambda = \ln(2)/22.3 \text{ yr} = 0.03108 \text{ y}^{-1}$) and t is age (y). ^{210}Pb flux is obtained by:

$$^{210}\text{Pb flux} = \Sigma^{210}\text{Pb}(\infty) \times \lambda \quad (2)$$

Major and trace element concentrations

For ICP-MS (inductively coupled plasma mass spectrometry) measurement of elemental concentrations, 0.25 g ash samples were digested in trace metal grade acids to extract trace metals. The HNO_3 -HF digestion method was applied after modification from Yafa and Farmer, (2006). Nine mL concentrated HNO_3 and 2mL diluted ($\text{HF}:\text{H}_2\text{O} = 1:1$) HF were added to ash samples in covered PFA beakers. One hundred μg Tl was also added into the solutions before digestion as an internal standard for measuring the loss of

elements during the whole operation. The samples were heated at least overnight at 150°C. After the solids were completely dissolved, the samples were evaporated down to 1 mL or less. The residue was dissolved into 20 mL 2% HNO₃ by heating. All the solutions were filtered (Whatman 542) and diluted into 50 mL polypropylene flasks. Solutions were filtered (0.2 µm polypropylene) and further diluted, as necessary, before concentrations were determined by ICP-MS (Varian 820-MS, Department of Chemistry). Indium was used as the instrumental internal standard. A total of 25 elements were assessed including five major elements (Al, Ti, Mn, Fe, Mg) and 20 trace elements (Cu, Zn, Sr, Cd, As, Sn, Sb, V, Pb, Bi, Ni, Zr, Rb, La, Ce, Pr, Nd, Sm, Th and U). Due to potential spectral overlap from $^{40}\text{Ar}^{35}\text{Cl}^+$, the As measurements may not be accurate (Olesik, 2000). The major and trace element test results were corrected by internal standard Tl and background samples, and calculated as both total peat and ash concentrations. All the elements concentrations in total peat were used to examine the correlations among the elements and between the elements with ash content in the pocosin by using SAS 9.3 program.

Pb and Sr isotopes

The isotopic composition of Sr (^{87}Sr , ^{86}Sr) and Pb (^{204}Pb , ^{206}Pb , ^{207}Pb and ^{208}Pb) were determined by thermal ionization mass spectrometry (TIMS) in the Department of Geological Sciences. Peat ash (20-50 mg) was digested in 1 mL concentrated HF and 1 mL 8M HCl on a hotplate at 150 °C for two hours, then evaporated to dryness. The residue was dissolved in 3mL 8.8M HNO₃ and 3 mL 8M HCl on a hotplate overnight in 15 mL PFA bottles. The samples were evaporated again to dryness, and the residue was

dissolved in 2 mL 0.5M HCl. Solutions were separated into two parts for Sr and Pb isotopes.

For Sr chemistry, the separated solution was dried and dissolved in 1 mL 3.5N HNO₃, then centrifuged. Eichrom SR-B100-S Sr Resin (about 200 µL) was used to collect Sr from the supernate. Before loading the sample, the resin was washed fully two times with MQ water (water that is purified through ion exchange) and 3.5N HNO₃. After loading the samples, they were rinsed multiple times with 3.5N HNO₃ with a total volume of 1.84 mL. Each rinse was started by adding three 30 uL drops, followed by performing five consecutive additions of one 350 µL aliquot each. The Sr was collected in beakers by rinsing two times with 500 µL MQ water.

The separated HCl solutions for Pb isotopes were dried and re-dissolved in 525 µl 1.1N HBr. Pb isotopes were collected in about 200 µL Eichrom anion resin AG1×8. Before loading the samples, the resin columns were rinsed two times with 500 µl 6N HCl, 500 µl MQ water and equilibrated by 250 µl HBr. The sample were rinsed by single drop 1.1N HBr (five times), 500 µl HBr and 250 µl 2N HCl (one time bulk rinse), and Pb was eluted with 500 µl 6N HCl. The process was repeated for collecting maximum Pb in samples. One drop of H₃PO₄ was added to both the final Sr and Pb solutions, which were then dried completely. The residue of Sr was re-dissolved in 2 µl 2M HCl and 2 µl dilute solution of TaCl₅, and the residue of Pb was re-dissolved in 3 µl silica gel for loading on filaments for mass spectrometry.

Results

Bulk properties

Bulk density increases rapidly with depth within the top 3 cm of the peat and stays around an average of 292 kg m^{-3} below 3 cm (Figure 5). Bulk density below the depth 17 cm is higher and more consistent compared to the values at shallower depths. The ash content of Core 1 is plotted as Loss on Ignition (Figure 5), and shows that within 10-30 cm depth, ash content is 5% on average. However, below 30 cm, ash material of Core 1 significantly increases and exceeds 20% of the total peat. The average water content is 70.7% throughout the whole core, which is lower than the observation in Ingram's (1987) study with an 80% average in the top 1.2 m.

Radiochemistry and peat chronology

The average supported ^{210}Pb concentration is $9.8 \pm 1.2 \text{ Bq kg}^{-1}$, and the total excess ^{210}Pb inventory is $2828 \pm 51 \text{ Bq m}^{-2}$ in the CNF pocosin. In the unsupported ^{210}Pb profile, the trend decreases from the surface to the depth of 12 cm in Core 1 (Figure 6). Two major peaks are shown in the main profile: one has the maximum activity ($303 \pm 9 \text{ Bq kg}^{-1}$) at 2.25 cm depth, and the other one is $76.5 \pm 4.6 \text{ Bq kg}^{-1}$ at 8 cm. Unsupported ^{210}Pb is taken as finite when its activity exceeds zero by two standard deviations. Below the main profile, finite excess ^{210}Pb are observed at the depth of 20 cm and 30 cm. The ^{137}Cs activity profile also displays two peaks at the same two depths as the ^{210}Pb maxima

(Figure 6). Finite ^{137}Cs exists through the whole core (i.e., ^{137}Cs exceeds zero by two standard deviations in all samples). The maximum value of ^{137}Cs activity is near the top of the core (2.25 cm depth) with $63.7 \pm 1.5 \text{ Bq kg}^{-1}$ and the other peak of $16.7 \pm 0.8 \text{ Bq kg}^{-1}$ is at 9 cm in Core 1. Complete radiochemistry results for cores 1 and 2 are presented in tabular form in the Appendices.

Based on the CRS model calculation, Core 1 provided about 120 years of peat depositional records between the surface and the depth of 20 cm (Figure 7). The peak of ^{137}Cs activities at depth interval 8 – 9 cm corresponds with the year interval 1959 – 1969 (Figure 6, 7). Peat Accumulation Rate (PAR) varies from $0.01 - 0.09 \text{ g cm}^{-2}\text{y}^{-1}$. The atmospheric ^{210}Pb flux in CNF pocosin was $88 \text{ Bq m}^{-2}\text{y}^{-1}$ (Table 2). Samples from two salt marshes near the study site on the coastal plain, yielded inventories of excess ^{210}Pb of 4220 Bq m^{-2} (flux $131 \text{ Bq m}^{-2}\text{y}^{-1}$) and 4560 Bq m^{-2} ($142 \text{ Bq m}^{-2}\text{y}^{-1}$) (Benninger and Wells, 1993). From Graustein and Turekian (1986), inventories of excess ^{210}Pb in undisturbed soil profiles were 4483 Bq m^{-2} (flux $139 \text{ Bq m}^{-2}\text{y}^{-1}$) in Raleigh, North Carolina and $3200\text{--}6333 \text{ Bq m}^{-2}$ (flux $99\text{--}197 \text{ Bq m}^{-2}\text{y}^{-1}$) in the Eastern U.S. Similar atmospheric deposition data from the peat bogs in the Western Europe are also shown in Table 2. The peat accumulation rate in this study is similar to the others in Table 2, but the ^{210}Pb inventory and therefore ^{210}Pb flux are lower than elsewhere in the Eastern U.S., except for some of the Florida marsh cores in Brenner et al. (2001). Both ^{210}Pb flux and inventory for these sites are lower than all the other records from North Carolina, and most of the other sites in Table 2. Meanwhile, the ^{137}Cs inventory of this study (865 Bq m^{-2}) is much lower than the average ^{137}Cs inventory (1947 Bq m^{-2} after decay correction to March 12th, 2011) in the Eastern and Mid-western U.S. (Graustein and Turkian, 1986).

Major elements abundance

Among the major elements (Al, Ti, Fe, Mg), Al and Ti have higher concentrations in peat above 10 cm and below 30 cm depth, which match the higher ash content deposition within the same depths through the core (Figure 8). The correlation table also shows strong correlations between Al, Ti and ash content (Table 3). Fe concentrations above 10 cm depth also increase with higher ash content but show no significant increasing trend below the depth of 30 cm. Mg concentration does not correlate with ash content (Table 3), and slowly increases from the bottom to the top of the core (Figure 8). The profiles of the major element Al, Ti and Fe accumulation rates have similar trends in the top 20 cm of the peat core (Figure 9), and it shows that between 1930 – 1984, Al, Ti, Fe accumulation rates slowly increased.

Trace elements abundance

The trace element concentrations in both peat and ash were obtained (Table 4 and 5). All trace elements except Zn and Sr have significant peaks in ash concentrations at the depth interval 12-13 cm (time interval 1930 – 1938). Excess ^{210}Pb is finite at 29-30 cm depth, and the concentrations of many of the trace elements in ash have local maxima near this depth.

All the elements can be separated into two groups: the elements Al, Ti, Mn, V, Cu, Zr, Cd, Sn, La, Ce, Pr, Nd, Sm, Th and U that correlate strongly (correlation coefficient is greater than 0.6) with ash content, and the elements Mg, Fe, Ni, Zn, Rb, As, Sb, Sr, Pb and Bi that are not strongly correlated with ash content (Table 3). Although the As measurement on ICP-MS could be inaccurate, As concentrations correlate strongly with

concentrations of other trace elements (Ni, Sb, Pb and Bi). In general, all the trace element concentrations in total peat increase when the ash content is high (Table 4). Several trace elements such as Pb, Sb, Bi and Ni have the maximum concentrations at depths of 8-9 cm (Figure 10), where the ash content is only moderately high. These elements are also highly correlated with each other and quite highly correlated with Fe (Table 3).

The trace elements Zr, La, Ce, Pr, Nd, Sm and Th have very low geochemical mobility and relatively low anthropogenic use compared with other elements such as Al, Fe, Cu, Zn, Pb, etc. Those elements all have high correlation with ash content as well as some major elements (Al, Ti, Mn) (Table 3). At the bottom of the core, REE concentrations are much higher than the average values.

Accumulation rate and enrichment factor of trace elements

The accumulation rates of all the trace elements were computed by applying the CRS ^{210}Pb dating model for the secure chronology in the last 100 years. The enrichment factor (E.F.) of all the trace elements was also produced and calculated by Ti and its crustal background (Shotyk et al., 2002):

$$\text{E.F.} = (\text{Conc. X} / \text{Conc. Ti})_{\text{sample}} / (\text{Conc. X} / \text{Conc. Ti})_{\text{UCC background}}$$

where X is a trace element; Conc. is mass-based concentration; and UCC is upper continental crust. UCC concentrations for selected trace elements X and Ti are the recommended values from Rudnick and Gao (2004). Results of this calculation show that most of the elements E.F. values are lower than 1.0; exceptions are Mg, Pb, Cd and Bi (Table 6). All the elements except Mn display trends with significant increases in E.F.

towards the core top, and they show three obvious peaks in their E.F. profiles at the depth intervals 12-13 cm, 28-29 cm and 38-39 cm, although three elements are significantly enriched in the samples Pb×4, Cd×20 and Bi×50 at their maximum values.

Pb and Sr isotopes

Pb and Sr isotopic compositions are shown in Table 7. The ratios of $^{206}\text{Pb}/^{207}\text{Pb}$, $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ display relatively constant values above the depth of 30 cm, and all the ratios increase dramatically below 30 cm of the core (Figure 11). Previous studies from other locations in the southeastern United States show similar $^{206}\text{Pb}/^{207}\text{Pb}$ ratios (about 1.20) from 1970 to present and lower $^{206}\text{Pb}/^{207}\text{Pb}$ ratios (about 1.19) between 1900 and 1970 (Jackson et al., 2004; Kamenov et al., 2009). Bollhöfer and Rosman (2001) summarized the range of $^{206}\text{Pb}/^{207}\text{Pb}$ isotope ratios from 75 studies in the Northern Hemisphere (United States, 1.159 – 1.231; Western Europe, 1.097 – 1.165; Asia, 1.091 – 1.177).

The $^{86}\text{Sr}/^{87}\text{Sr}$ profile is shown in Figure 12 with Sr concentration in ash and Sr E.F.. The $^{86}\text{Sr}/^{87}\text{Sr}$ ratio has the highest value at the bottom of the core and decreases when the peat becomes shallower. When the depth reaches 12 cm, the $^{86}\text{Sr}/^{87}\text{Sr}$ ratio is the lowest. Then it starts to increase again towards the surface of the core.

Discussion

Pocosin ombrotrophic and minerotrophic characteristics

At the bottom of the core (below 30 cm), the significantly higher ash content may reflect the transitional character from a mineralogical fen to an ombrotrophic bog. The high organic content shows the ombrotrophic character of the pocosin between depths of 10-30 cm. Above 10 cm, high ash content (greater than 10%) (Figure 5) implies that the peat in the study site has accepted significant soil-dust deposition. The high ash content in this top layer which represents the past 50 years may relate to the frequent forest fires in the CNF pocosin. Below 30 cm of the core, the extensive ash content with higher Al, Ti, Zr and REE concentrations reveals that the materials are most likely derived from the underlying regolith, implying a minerotrophic character. Additionally, from the contour map of peat thickness reported in Ingram (1999), the range of peat thickness around the study site is about 0-1 m deep. It suggests that the source of the bottom material of the core is possibly from underlying soil or bedrock.

Sediment chronology

Previous studies rely on chronology obtained by using both ^{210}Pb and ^{137}Cs . Studies have shown that the immobility of ^{210}Pb makes the chronology results agree with the ^{14}C dating results (Shotyk et al., 1998), and that ^{137}Cs is often used as the sole marker of chronology (Oldfield et al., 1979; MacKenzie et al., 1997). However, Urban et al. (1990)

proposed that ^{210}Pb was mobile under the water table. In that case, the part of peat preserved under the water table would contain less ^{210}Pb deposition, jeopardizing the chronology. Thus, the problem is more significant where the water table is shallow (beneath hollows) than where it is deeper (beneath hummocks). In consequence, lower ^{210}Pb inventory and younger age from ^{210}Pb dating were often observed in the cores that were collected from hollows in peat bogs. Although we did not measure the water table of the study site, due to the facts that the water table was shallow during the field sampling and the nearby ditch was also shallow, the water table in the CNF pocosin should be shallow. However, at the study site, no obvious surface elevation changes were observed. Therefore, whether ^{210}Pb activities near the top of the core could be affected by the water table in this study needs more evidence to further advance this discussion.

There are two unexpected small peaks at approximately 20 cm and 30 cm in the ^{210}Pb and ^{137}Cs activities profiles. They indicate vertical transportations that took more recent ^{210}Pb and ^{137}Cs deposition into the deeper peat during the last century. Since there were no visible live animals or burrows throughout the whole core, bioturbation is not a likely explanation.

From the chronology obtained by ^{210}Pb dating, the peak of ^{137}Cs activity at the depth of 9 cm correlates with the peak deposition of nuclear weapons fallout in 1963. The maximum ^{137}Cs activity near the surface of the core may be caused by the absorption from the upper plants on the peat (Oldfield et al., 1979). Different location trends of ^{137}Cs profiles in Oldfield et al., (1979) were also explained as the effect of predominance of different plant species. The dominant flora of the North Carolina pocosin in this study are mostly a mix of scrub-shrub and pond-pine (Richardson, 2003), which differ from the

vegetation (*Eriophorum vaginatum* with rich roots) in Oldfield et al., (1979). The abundant leaves and a few thin roots near the top core samples suggest the possibility of the upper plants effects on ^{137}Cs in this study.

Similar ^{137}Cs trends were also found in other studies. Olid et al., (2010) discussed profiles of ^{137}Cs and ^{241}Am in peat from the NW Iberian Peninsula, in which both ^{137}Cs and ^{241}Am have peak activities around 1963 at three out of the four study sites, and the maximum activities of ^{137}Cs also appear to be near the surface at those study sites. However, studies from the west of Ireland (Coggins et al., 2006) and the Great Hinggan Mountains in NE China (Bao et al., 2009) only observed one peak from the ^{137}Cs concentration profiles corresponding to 1963, from the fallout deposition of atmospheric testing of nuclear weapons.

Major elements, Zr and REE

Al, Mg, Ti, Mn, Zr and REE were tested to track the characteristics of the original soil/dust source or mineral mines inputs that influenced the pocosin (Weiss et al., 1999; Shotyk et al., 2002b). In CNF pocosin, Al, Ti, Mn, Zr and REE concentrations in peat strongly correlate with ash content through the whole core (Table 3), which reveals that their concentrations increase with enhanced mineral deposition. However, Fe concentrations do not correlate with Al and Ti, but highly correlate with Pb in the core (Table 3). The reason for the high correlation between Pb and Fe remains unclear.

The lithogenic elements such Ti and Zr show conservative behavior during weathering process of rocks, meaning that they were enriched in soil/mineral dust (Shotyk et al., 2001; 2002). The very strong correlation between Ti and Zr ($r = 0.992$) in

Table 3 suggests that either of them can be used as an index to present the original mineral dust input in the peat, as well as a factor in the background of enrichment factor calculation.

Mineral sources that are high in silica can result in increasing Al and Fe levels in peat. They can be from the mining of non-metal minerals, such as crushed stone and construction sand and gravel, which are widely distributed in the coastal plain of North Carolina. Raven and Loeppert (1997) found abundant Fe, Mn and Ti in rock phosphate samples. Therefore, the mining process of phosphate in Beaufort County (about 80 km north of CNF, Figure 1) could have introduced Fe, Mn and Ti to the study site over the last 60 years (Stuckey, 1965). Additionally, more activities such as the deforestation and agriculture expansion that introduce enhance dust deposition could also have increased these elements concentrations.

The nature of the mixed-vegetation (pine tree and shrub) in the Croatan National Forest (CNF) makes it difficult to approach the study site. Therefore, the study site was chosen close to secondary gravel roads and small ditches (Figure 4). This location raises the issue that the process of building roads could have introduced dust to the site. The material of the road is mostly limestone, and it may contain silicate sand. Thus, crushed stone mines, construction sand and gravel, phosphate mines, local farming and road construction could have been the primary external sources of dust to the CNF pocosin.

Trace element deposition in CNF pocosin

Among all the trace elements that are tested in this study, the group 1 (V, Cu, Zr, Cd, Sn, La, Ce, Pr, Nd, Sm, Th and U) elements that have higher correlation with ash content

were probably mainly introduced from mineral dust, whereas the group 2 (Ni, Zn, Rb, As, Sb, Sr, Pb and Bi) ones that have relatively weak correlation with ash content may have also been introduced by other anthropogenic sources (Table 3). In the last century, most trace elements concentrations in ash start to increase from the depth 17 cm to 13 cm, corresponding with the year from 1905 to 1930. This may represent the period of the beginning of North Carolina industrialization. At these same depths, however, their concentrations in the total peat do not have a peak, which is due to the low ash content (less than 5%).

The enrichment factor (E.F.) of trace elements is used to distinguish the earth material background from anthropogenic input (Shotyk, 2002a). The higher E.F. of certain element(s) can be caused by extensive anthropogenic input. From the results of the E.F. calculation in this study, a number of trace elements have E.F. values lower than 1.0 (Table 6), while their profiles still display obvious variations. Most of the trace elements E.F. above 16 cm (after the year of 1893) are higher than those values at the bottom core. This suggests the significant anthropogenic source input in the last 120 or so years. Enrichment factors show that the soil material in the CNF demonstrates a localized and thin upper continental crust layer, instead of the average upper continental crust in North America (Rudnick and Gao, 2004).

Trace elements group one

Group 1 elements (V, Cu, Zr, Cd, Sn, La, Ce, Pr, Nd, Sm, Th and U) were probably introduced from sources that released significant mineral dust into the atmosphere such as crush stone mines, construction sand and gravel, phosphate mines and agricultural soil in

the coastal plain of North Carolina. The strong correlation between these elements and major elements Ti and Al in peat indicate that these trace elements were deposited in proportion to the input of weathered minerals. Pocosin forest fires can also significantly affect trace elements concentrations in peat. When they occurred, parts of vegetation and peat were turned into ash, resulting in a thinner layer of peat with higher ash content.

Cu is one of the most commonly reported trace elements (Pb, Cu, Zn) in studies of atmospheric deposition. In southeastern Florida, the Cu trace element concentrations are close to the levels shown in this study (Kamenov et al., 2009). The comparisons in Figure 13 reveal Cu concentrations trends are different between the two studies, showing no increasing trend near the surface of Core 1 from CNF pocosin. The comparison may imply the difference in Cu usage in the recent years between Florida and North Carolina. In New York City, Cochran et al., (1998) observed that the two salt marshes have high Cu concentrations in ash, which are about six times higher than the records in CNF pocosin (Figure 14), suggesting greater Cu pollution in NYC.

The Cd maximum concentration in the CNF pocosin is 1.18 ppm at depths of 34-35 cm (Table 5, Figure 13) which is much higher than the maximum Cd concentration in the Florida peat sediment (0.072 ppm) (Kamenov et al., 2009). Cd concentration (ash) in North Carolina is also higher than the ones in New York City (Figure 14). The possible sources of Cd in North Carolina are the nearby phosphate mines, and coal-fired power plants fly ash (Querol et al, 1999).

Raven and Loeppert, (1997) summarized that phosphate rocks and phosphate fertilizer can introduce a number of trace elements to the environment. The comparison

between the trace elements in North Carolina phosphate rock (Raven and Loeppert, 1997) and that in CNF pocosin, it is possible that Cu, Zn, Rb, Sr, Zr, Cd and U elements are introduced from dust suspended during phosphate mining.

Trace elements group two

The other group elements (Ni, Zn, Rb, As, Sb, Sr, Pb and Bi) have weak correlations with ash content that may be influenced by other different anthropogenic sources such as leaded gasoline usage and coal combustion during the last century. These records can also be affected by the local population growth rate and the distances from the coal-fired power plants to the study site.

Different Pb records in the CNF (Figure 15) indicate that the industrialization and the introduction of leaded gasoline could have dominated the Pb sources during the period between 1908 and 1933. After 1933, Pb accumulation rate rapidly increased until the year 1970, which coincides with a period of growing usage of leaded gasoline in North Carolina. Therefore, leaded gasoline usage may be one of the dominating Pb sources in the area during this period. Moreover, Pb emissions are also affected by the growth rate of populations in the surrounding counties, which correlate with the number of leaded gasoline-using vehicles on roads. Within the five adjacent counties, the highest population growth rates were found in the 1950s and 1960s, which match the rapid Pb accumulation rate during the same period (Figure 15). Between 1970 and 1983, the Pb concentrations were lower than the previous period in both peat and ash, and Pb accumulation rate did not accelerate as fast as before. This period matches the beginning of leaded gasoline phasing-out at 1973. The remaining high level of Pb concentrations

may be related to other Pb sources such as the coal combustion in the area, phosphate mines in Beaufort county and dust from phosphate fertilizer (Raven and Loeppert, 1997).

The comparisons of Pb between different regions in the United States are shown in Figure 13 and 14. Both Florida (Kamenov et al. 2009) and CNF peat contain Pb maxima at depths corresponding to 1970. It suggests the similarity in leaded gasoline usage period between Florida and North Carolina. Cochran et al., (1998) observed that the two salt marshes in New York City have high fluxes of Pb in ash. Comparison shows their concentrations in ash are several times higher ($Pb \times 9$) than the records in the CNF pocosin (Figure 14), which implies greater trace metal pollution in cities such as New York City. The same comparisons for Zn also indicated the similar Zn maxima nearer the core tops in Florida, while a higher concentration was observed in NYC ($Zn \times 5$) relative to that in North Carolina. This supports the notion that extra Pb can be from the emissions from motor vehicles (Huang et al, 1994), and that more Pb and Zn can be released to the atmosphere near a denser populations and heavier-traffic areas.

To compare Pb concentrations in the U.S. with those in Europe, numerous studies have shown that the highest Pb concentration in peat is at the depth associated with years around 1970 (Shotyk et al., 1995, 1998; Weiss et al., 1999; Cloy et al., 2008; Novak et al., 2008). However, from the studies in Northeast China, the highest Pb concentration was found around 2000, when leaded gasoline began to be phased out (Bao et al., 2010). Therefore, the concentrations within peat of trace elements such as Pb can show both similarities and differences in local industrial development at regions within the U.S., as well as among countries.

The very strong correlation (r is great than 0.8, Table 3) between Pb concentration and some other elements (Ni, As, Sb, Bi) concentrations suggest they were probably introduced from similar anthropogenic sources. Sb, Bi and Ni all have their highest concentrations (in peat) at depths of 8–9 cm (time interval 1959 – 1969) (Table 4, Figure 10). The excess Sb and Bi are most likely from anthropogenic input since their concentrations are very low in fresh water and they are relatively volatile (Kamenov et al., 2009). The high Sb, Bi and Ni concentrations between 1960 and 1975 in CNF pocosin may be from motor vehicle emissions (Huang et al., 1994). Previous studies have shown that fossil fuel combustion, coal mining and fly ash from coal-fired power plants often carry extra Sb, Bi and Ni into the atmosphere (Kaakinen et al., 1975; Querol et al, 1999; Kamenov et al., 2009). Coal burning has constituted a large fraction of the energy generation of North Carolina, although coal mining has not been significant. Therefore, coal combustion could be one of the Sb, Bi and Ni sources in CNF pocosin.

In general, the elements (V, Cu, Zr, Cd, Sn, La, Ce, Pr, Nd, Sm, Th and U) that strongly correlate with ash content were possibly introduced by the dust from phosphate mines, agricultural soil, crushed stone mines and fly ash from coal combustion. The other group of elements (Ni, Zn, Rb, As, Sb, Sr, Pb and Bi) that do not show strong correlation with ash content may have been mainly emitted by motor vehicles, particularly cars that used leaded gasoline, and also by coal combustion.

Sources to the CNF pocosin based on Sr and Pb isotopes

Based on Faure's (1986) model, the $^{87}\text{Sr}/^{86}\text{Sr}$ in oceans is defined as a combination of different strontium sources including weathering young volcanic rocks (0.704 ± 0.002),

old sialic rock (0.720 ± 0.005) and marine carbonate rocks (0.708 ± 0.001). The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio in the modern seawater is 0.70918 (Frank, 2002), whereas the average $^{87}\text{Sr}/^{86}\text{Sr}$ ratio in CNF pocosin is 0.71361 ± 0.00047 . The high $^{87}\text{Sr}/^{86}\text{Sr}$ ratio (above 0.71196) throughout the whole core implies that the radiogenic Sr sources dominate the deposition in the CNF. The bedrock of CNF pocosin is fossiliferous clay and sand, shelly sand, sandy marl and limestone that formed in the Tertiary. The high $^{87}\text{Sr}/^{86}\text{Sr}$ ratio in the bottom core (below 30 cm) also implies that the underlying rock may contain more sialic rocks which usually have a higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratio. The less radiogenic Sr sources at depth of 12 cm suggest that the additional materials such as marine aerosols with low $^{87}\text{Sr}/^{86}\text{Sr}$ ratio character were deposited in the CNF pocosin around 1940's. Tobiassen (1982) measured the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio (0.70790-0.70935) in phosphate grains from the Miocene Pungo River formation in North Carolina. Therefore a marine isotopic signature can be introduced from anthropogenic activities such as phosphate mining and crushed stone (shell limestone) mining in coastal plain during the last 100 years.

The Pb isotope ratios ($^{206}\text{Pb}/^{204}\text{Pb}$, $^{206}\text{Pb}/^{207}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$, $^{208}\text{Pb}/^{204}\text{Pb}$) do not have any significant variations in the top 30 cm of Pb deposition (Table 8, Figure 16). This suggests no obvious changes in the sources of Pb to the CNF pocosin during the last century. The anthropogenic Pb sources can be from leaded gasoline usage in North Carolina. The $^{206}\text{Pb}/^{207}\text{Pb}$ ratio (1.1992 – 1.2366) is close to the values in other areas in U.S. such as Tampa (1.214 ± 0.001 , 1.208-1.231), Argonne, France (1.211 ± 0.001), and New York (1.197-1.223) (Bollhöfer and Rosman, 2001). Kamenov et al., (2009) claimed that Florida has been using different source ore for Pb and higher $^{206}\text{Pb}/^{207}\text{Pb}$ ratio (above 1.200), representing more radiogenic Mississippi Valley Type (MVT) ore in the last 40

years. Similarly high $^{206}\text{Pb}/^{207}\text{Pb}$ ratios were found in Georgia (Jackson et al., 2004) for the last 50 years and in the Chesapeake Bay area (Marcantonio et al., 2002) for the last 35 years.

Coal combustion has also been claimed to be an important source of Pb in North America, Western Europe and East Asia (Bollhöfer and Rosman, 2001; Diaz-Somoano et al., 2009). Diaz-Somoano et al., (2009) observed that the $^{206}\text{Pb}/^{207}\text{Pb}$ in coal from the U.S. ranges from 1.1907 – 1.2314 and that $^{208}\text{Pb}/^{206}\text{Pb}$ ranges from 2.0201 – 2.0829. These Pb isotopic records in coal are comparable with those in CNF pocosin ($^{206}\text{Pb}/^{207}\text{Pb}$: 1.1992 – 1.2366; $^{208}\text{Pb}/^{206}\text{Pb}$: 2.0479 – 2.0851). Therefore, coal combustion could also be one of the major Pb sources in North Carolina during the last 100 years.

Figure 17 shows the comparison of Pb isotopic compositions ($^{207}\text{Pb}/^{206}\text{Pb}$ and $^{208}\text{Pb}/^{206}\text{Pb}$) between CNF pocosin in North Carolina and other study sites in the eastern United States. The data that lay in the overlapping area in the plot are the samples from CNF pocosin Pb deposition in the last 100 years, Florida swamp in the last 40 years and the northeastern U.S. Lake Andrus in the last 40 years (Graney et al., 1995). This demonstrates that in over the last 40 years, the North Carolina coastal plain was characterized by Pb sources similar to those of other places in the eastern U.S.

Conclusion

The Croatan National Forest pocosin, even though it has both ombrotrophic and minerotrophic characteristics, can be used as an archive of the atmospheric deposition of trace elements such as lead. The distribution of pocosin peat in North Carolina offers many opportunities to expand this study to different locations in the coastal plain and to conduct more detailed analysis with longer sample cores, which will provide more information about the local history.

By using both ^{210}Pb and fallout radionuclides, along with trace elements concentrations and Pb isotopes, a robust chronology was constructed. Mobility of ^{210}Pb , Pb and other trace elements concentration is possible due to the water table influence on the peat; however, further work is would be needed to better evaluate that possibility. Additionally, the ^{137}Cs was moving chemically in the peat, independent of the effects of plants. Therefore, additional analysis testing immobile radionuclides such as ^{242}Pu or ^{241}Am could help to better define the peat chronology.

The recent lead input reflects the local/regional lead emissions from industrial activities in the coastal plain of North Carolina. From the late 1800's to the early 1900's, local industrialization, along with development related to World War II, expanded local farming, mines and roads. Half a century later, when leaded gasoline was phased out in 1973, the trace elements levels in the atmosphere started to drop. Moreover, the record of other trace elements as well as some major elements represents more local history such as

phosphate mines, crushed stone mines, forest fires, road construction and coal combustion. Historical (100 years) Pb deposition in North Carolina was similar to that observed in Europe, where industrial development has been similar, but different from the patterns observed in China.

Table 1 North Carolina local anthropogenic history.

Time Period	Historical Events
1500s to 1700s	Early European Settlers, fire for agriculture purpose in Southern U.S. ¹
1800 to mid-1800's	Gold mines started and the most significant production period in North Carolina. ²
1890s to 1920s	Industrialization; initiation of a variety of mines (i.e. gold mines); the rapid growth in the timber industry and associated logging led to increasingly numerous, intense and widespread fires. Many Coastal Plain forests were slashed-and-burned, or often accidentally ignited, causing more severe fires, especially for 12 NC counties in 1894. ^{1,2}
1905	The first production of sand and gravel, crushed stone in the coastal Plain of North Carolina. ^{2,3}
1920s to 1940s	In the southern U.S., fire suppression (not allowing fires) was sponsored in this period, which allowed Appalachian hardwood forests with other species, replacing many areas of oak/pine fire-tolerant species. ¹
1923	Introduction of leaded gasoline in U.S. ⁴
1950s	The highest population change/growth rate (53.1%) in the adjacent counties (Figure 2, 3).
1951-1958	The first phosphate mines in Beaufort county. ²
1960s	The second highest population change/growth rate (43.2%) in the adjacent counties (Figure 2, 3); Renewed phosphate mines in Beaufort county. ²
1973	Leaded gasoline phaseout began ⁴
1990s	Regular wildland fire by forest fire management in the coastal plain. ¹

¹Fowler and Konopik, 2007; ² Stuckey, 1965; ³ NCGS: <http://www.geology.enr.state.nc.us/Mineral>; ⁴ US EPA.

Table 2 Peat accumulation rate, ^{210}Pb inventory and atmospheric ^{210}Pb flux comparisons from different atmospheric deposition sites.

Country	Region/Site	Peat accumulation rate ($\text{g cm}^{-2}\text{yr}^{-1}$)	^{210}Pb inventory (Bq m^{-2})	Atmospheric ^{210}Pb flux ($\text{Bq m}^{-2}\text{yr}^{-1}$)	References
Switzerland	Tramelan, bogs	0.02-0.04	4690	146	Appleby et al., (1997)
UK	Glasgow, peat bog	0.027-0.040	3380-3500	106-109 ^a	Mackenzie et al., (1998)
NW Iberian	Galicia, mires	-	4994-10135	155-315 ^a	Olid et al.,(2010)
United States	North Carolina, pocosin	0.01-0.09	2829	88	This study
	North Carolina, salt marshes	-	4220, 4560	131, 142	Benninger and Wells, (1993)
	Florida, marshes	0.0035 (ash)	3980-7585	124-236 ^a	Kamenov et al., (1997)
	Florida, marshes		1916-3966	60-123 ^a	Brenner et al., (2001)
	New York, bog	-	2667-9000	83-279 ^a	Cochran et al., (1997)
	New York, salt marsh	0.02 ^a , 0.0017(ash) ^a	3404	106	Urban et al., (1990)
	East U.S., undisturbed soil	-	3200-6333	99-197 ^a	Graustein and Turkian, (1986)
	Raleigh, NC		4483	139 ^a	
	Bluff Mountain, NC		5567	173 ^a	

^a Calculated value using 22.3 yr as the half-life of Pb.

Table 3 Elements concentrations correlation relationship table. Dark grey represents very strongly correlated elements (Correlation Coefficient > 0.8); Light grey represents strong correlated elements relationship (Correlation Coefficient is from 0.8-0.6). r is considered statistically significant when $p < 0.05$.

[illegible]

Table 4 Trace elements concentration in peat (uncertainties estimated at $\leq 10\%$) from Croatan National Forest pocosin, NC, U.S.

Depth cm	V ppm	Ni ppm	Cu ppm	Zn ppm	Rb ppm	Sr ppm	Zr ppm	Cd ppm	As ppm	Sn ppm	Sb ppm	La ppm	Ce ppm	Pr ppm	Nd ppm	Sm ppm	Pb ppm	Bi ppm	Th ppm	U ppm	LOI %
0.5-2.25	9.46	2.79	5.95	11.6	5.41	33.8	12.5	0.425	0.914	0.480	0.287	4.83	7.99	1.10	3.93	0.689	9.09	0.908	1.24	0.457	85.9
4-5	15.0	4.86	5.10	15.3	3.91	27.7	16.2	0.526	1.88	0.814	0.340	5.94	10.9	1.38	4.95	0.867	15.2	1.61	1.69	0.793	74.5
6-7	7.17	3.57	3.70	5.22	2.07	11.0	8.27	0.348	1.30	0.597	0.293	4.27	6.78	0.943	3.33	0.580	13.1	1.34	1.05	0.514	89.2
8-9	13.3	5.21	6.06	6.87	1.80	13.3	13.7	0.513	2.02	1.12	0.544	6.05	10.1	1.36	4.93	0.903	25.3	2.53	1.63	0.853	81
10-11	3.94	2.70	2.05	2.42	1.11	8.66	4.71	0.207	1.01	0.349	0.202	2.94	4.98	0.713	2.51	0.441	7.34	0.870	0.677	0.343	94.8
12-13	1.94	1.88	1.70	1.52	0.396	5.84	2.40	0.099	0.625	0.168	0.083	1.43	2.49	0.355	1.23	0.212	2.94	0.367	0.389	0.217	98.2
14-15	3.05	2.77	2.59	1.51	0.677	6.78	4.46	0.154	0.933	0.265	0.094	2.38	4.09	0.574	1.98	0.334	4.27	0.457	0.809	0.404	96.1
16-17	4.31	2.45	2.67	0.872	0.493	6.08	5.83	0.183	0.772	0.217	0.074	3.15	4.75	0.627	2.13	0.357	4.15	0.438	0.962	0.423	91
18-19	6.00	2.57	3.03	3.43	0.203	6.45	8.40	0.252	0.800	0.266	0.074	2.37	4.48	0.509	1.76	0.310	4.85	0.527	0.966	0.572	87.9
20-21	5.84	2.39	2.79	4.06	0.300	5.93	8.59	0.253	0.705	0.285	0.077	2.55	4.63	0.533	1.84	0.319	4.50	0.498	1.03	0.514	87.9
22-23	5.56	2.21	2.68	3.52	0.075	4.53	7.72	0.218	0.634	0.233	0.062	1.13	2.28	0.268	0.959	0.175	3.68	0.380	0.703	0.451	90.8
24-25	6.29	2.73	3.64	3.17	0.043	4.50	7.59	0.212	0.768	0.226	0.071	0.771	1.62	0.194	0.711	0.133	4.36	0.401	0.612	0.464	91.4
26-27	5.49	2.87	3.96	1.30	0.124	5.43	6.99	0.197	0.860	0.225	0.079	1.90	3.62	0.455	1.63	0.289	4.54	0.521	1.33	0.493	90.9
28-29	7.29	3.60	5.63	2.73	0.074	5.18	10.5	0.307	1.08	0.296	0.120	1.11	2.22	0.258	0.934	0.172	5.24	0.482	0.818	0.571	92.4
30-31	9.45	3.57	6.29	3.36	0.104	5.50	14.2	0.404	1.19	0.455	0.145	0.824	1.71	0.207	0.759	0.139	5.10	0.942	0.704	0.625	91
32-33	5.94	1.56	3.32	1.33	0.145	3.63	11.7	0.311	0.585	0.308	0.085	0.941	2.07	0.256	0.949	0.176	2.22	0.434	0.679	0.406	91.9
34-35	23.0	4.01	10.0	8.84	1.70	14.5	42.3	1.20	1.52	0.973	0.283	7.53	15.5	1.82	6.53	1.11	7.34	1.46	3.10	1.25	50.4
36-37	20.3	2.64	6.96	3.77	2.02	12.1	37.4	1.02	1.17	0.908	0.237	5.85	12.4	1.52	5.48	0.975	5.10	1.06	2.76	1.11	67.2
38-39	15.3	1.84	5.02	2.27	2.30	9.46	27.4	0.727	0.886	0.783	0.181	5.95	10.6	1.46	5.23	0.893	4.08	0.837	2.30	0.937	80.5
40-41	27.4	2.98	8.39	5.27	3.96	15.4	43.2	1.15	1.27	1.38	0.288	7.76	15.8	2.06	7.49	1.33	7.08	1.46	3.69	1.59	64.6
42-43	20.1	2.13	5.22	6.00	2.41	10.3	31.7	0.844	0.924	1.03	0.216	2.36	5.51	0.711	2.69	0.520	5.45	1.13	2.17	1.13	71.5

Table 5 Trace elements concentrations in ash (uncertainties estimated at $\leq 10\%$) from Croatan National Forest pocosin, NC, U.S.

Depth interval cm	V ppm	Ni ppm	Cu ppm	Zn ppm	Rb ppm	Sr ppm	Zr ppm	Cd ppm	As ppm	Sn ppm	Sb ppm	La ppm	Ce ppm	Pr ppm	Nd ppm	Sm ppm	Pb ppm	Bi ppm	Th ppm	U ppm
0.5-																				
2.25	67.1	19.8	42.3	82.3	38.4	240	88.5	3.02	6.49	3.41	2.04	34.3	56.8	7.81	27.9	4.90	64.5	6.45	8.80	3.24
4-5	58.7	19.0	20.0	59.8	15.3	109	63.6	2.06	7.36	3.19	1.33	23.3	42.6	5.39	19.4	3.40	59.5	6.33	6.62	3.11
6-7	66.7	33.2	34.4	48.5	19.2	102	76.9	3.23	12.1	5.55	2.72	39.7	63.1	8.77	30.9	5.39	122	12.4	9.73	4.78
8-9	69.6	27.4	31.8	36.1	9.47	69.8	71.7	2.70	10.6	5.86	2.86	31.8	53.3	7.16	25.9	4.74	133	13.3	8.55	4.48
10-11	76.1	52.2	39.6	46.7	21.4	167	91.1	4.01	19.5	6.74	3.90	56.7	96.3	13.8	48.5	8.53	142	16.8	13.1	6.62
12-13	105	102	92.4	82.4	21.5	317	130	5.37	33.9	9.15	4.53	77.9	135	19.3	67.0	11.5	160	19.9	21.1	11.8
14-15	78.2	70.9	66.4	38.7	17.4	174	114	3.94	23.9	6.79	2.40	61.0	105	14.7	50.8	8.58	110	11.7	20.8	10.4
16-17	47.7	27.1	29.6	9.66	5.46	67.3	64.6	2.02	8.55	2.40	0.814	34.9	52.6	6.95	23.6	3.96	45.9	4.86	10.7	4.69
18-19	49.6	21.2	25.1	28.4	1.68	53.3	69.5	2.08	6.62	2.20	0.609	19.6	37.0	4.21	14.6	2.56	40.2	4.36	7.99	4.73
20-21	48.2	19.7	23.0	33.5	2.47	48.9	70.9	2.08	5.82	2.35	0.635	21.0	38.2	4.40	15.2	2.63	37.1	4.11	8.53	4.24
22-23	60.2	23.9	29.0	38.1	0.811	49.1	83.7	2.36	6.87	2.52	0.671	12.2	24.7	2.91	10.4	1.89	39.9	4.12	7.61	4.89
24-25	72.9	31.6	42.1	36.7	0.500	52.2	87.9	2.46	8.90	2.62	0.825	8.94	18.7	2.24	8.24	1.54	50.5	4.65	7.09	5.37
26-27	60.6	31.7	43.7	14.4	1.37	59.8	77.1	2.18	9.48	2.48	0.876	21.0	39.9	5.02	18.0	3.19	50.0	5.74	14.6	5.44
28-29	95.5	47.2	73.8	35.8	0.969	67.9	138	4.02	14.1	3.88	1.58	14.6	29.1	3.38	12.2	2.25	68.7	6.32	10.7	7.48
30-31	105	39.6	69.7	37.3	1.16	61.0	158	4.48	13.1	5.05	1.61	9.14	18.9	2.30	8.43	1.55	56.5	10.5	7.81	6.93
32-33	73.7	19.3	41.1	16.5	1.79	45.0	145	3.86	7.26	3.82	1.06	11.7	25.7	3.17	11.8	2.18	27.5	5.38	8.41	5.03
34-35	46.3	8.08	20.2	17.8	3.44	29.3	85.3	2.42	3.07	1.96	0.572	15.2	31.3	3.67	13.2	2.25	14.8	2.94	6.25	2.52
36-37	61.9	8.06	21.2	11.5	6.16	37.0	114	3.10	3.58	2.77	0.722	17.9	37.7	4.63	16.7	2.98	15.5	3.23	8.44	3.38
38-39	78.5	9.43	25.7	11.6	11.8	48.4	140	3.72	4.54	4.01	0.929	30.4	54.1	7.48	26.8	4.57	20.9	4.28	11.8	4.79
40-41	77.4	8.42	23.7	14.9	11.2	43.4	122	3.24	3.59	3.89	0.813	21.9	44.5	5.81	21.2	3.75	20.0	4.12	10.4	4.48
42-43	70.6	7.45	18.3	21.0	8.43	36.1	111	2.96	3.24	3.62	0.76	8.25	19.3	2.49	9.44	1.82	19.1	3.94	7.61	3.97

Table 6 Enrichment factor (E.F.) of trace element in CNF pocosin, U.S.

Depth (cm)	Al	Mn	Fe	Mg	V	Ni	Cu	Zn	Rb	Sr	Zr	Cd
0-2.25	0.64	0.08	0.17	2.8	0.35	0.21	0.76	0.62	0.10	0.38	0.54	17
4-5	0.69	0.05	0.32	4.6	0.35	0.23	0.41	0.51	0.05	0.19	0.44	13
6-7	0.58	0.05	0.30	3.7	0.36	0.37	0.65	0.38	0.05	0.17	0.49	19
8-9	0.85	0.05	0.31	10	0.38	0.31	0.60	0.29	0.03	0.12	0.46	16
10-11	0.98	0.04	0.27	13	0.37	0.53	0.67	0.33	0.05	0.25	0.53	21
12-13	0.73	0.05	0.17	7.5	0.39	0.78	1.2	0.44	0.04	0.36	0.57	21
14-15	0.49	0.04	0.14	4.9	0.31	0.58	0.91	0.22	0.03	0.21	0.54	17
16-17	0.45	0.03	0.11	4.1	0.31	0.36	0.67	0.09	0.02	0.13	0.50	14
18-19	0.34	0.03	0.13	3.0	0.31	0.26	0.54	0.25	0.01	0.10	0.49	14
20-21	0.74	0.03	0.16	4.9	0.30	0.25	0.50	0.30	0.01	0.09	0.52	14
22-23	0.42	0.02	0.12	2.7	0.33	0.27	0.55	0.30	0.00	0.08	0.55	14
24-25	0.31	0.01	0.07	1.7	0.34	0.31	0.69	0.25	0.00	0.07	0.49	12
26-27	0.22	0.01	0.04	0.95	0.33	0.36	0.83	0.11	0.00	0.10	0.50	13
28-29	0.21	0.04	0.02	0.65	0.30	0.31	0.81	0.16	0.00	0.07	0.52	14
30-31	0.18	0.02	0.02	0.38	0.26	0.20	0.59	0.13	0.00	0.05	0.46	12
32-33	0.23	0.02	0.02	0.35	0.20	0.11	0.40	0.07	0.00	0.04	0.48	12
34-35	0.24	0.02	0.02	0.31	0.24	0.09	0.37	0.14	0.01	0.05	0.53	14
36-37	0.21	0.02	0.02	0.25	0.21	0.06	0.24	0.06	0.01	0.04	0.45	11
38-39	0.23	0.10	0.02	0.35	0.22	0.05	0.25	0.05	0.02	0.04	0.46	11
40-41	0.24	0.07	0.02	0.31	0.24	0.05	0.26	0.07	0.02	0.04	0.45	11
42-43	0.21	0.10	0.02	0.25	0.22	0.05	0.20	0.10	0.01	0.03	0.42	10

Depth (cm)	As	Sn	Sb	La	Ce	Pr	Nd	Sm	Pb	Bi	Th	U
0-2.25	0.68	0.82	2.6	0.56	0.45	0.55	0.52	0.53	1.9	20	0.42	0.61
4-5	0.88	0.87	1.9	0.40	0.36	0.40	0.38	0.38	2.0	23	0.36	0.66
6-7	1.3	1.4	3.6	0.68	0.53	0.66	0.61	0.61	3.8	41	0.49	0.93
8-9	1.2	1.5	3.8	0.49	0.40	0.48	0.46	0.48	4.2	44	0.43	0.88
10-11	1.9	1.5	4.6	0.70	0.59	0.75	0.69	0.70	4.0	50	0.59	1.2
12-13	2.5	1.6	4.1	1.6	1.4	1.7	1.6	1.5	3.4	45	0.72	1.6
14-15	1.9	1.2	2.3	1.1	0.96	1.2	1.1	1.1	2.5	28	0.76	1.5
16-17	1.1	0.72	1.3	0.68	0.50	0.59	0.53	0.51	1.7	19	0.64	1.1
18-19	0.79	0.63	0.91	0.34	0.31	0.32	0.29	0.29	1.4	16	0.44	1.0
20-21	0.73	0.67	0.96	0.36	0.32	0.33	0.30	0.30	1.3	15	0.49	0.95
22-23	0.76	0.64	0.90	0.12	0.12	0.13	0.12	0.12	1.3	14	0.39	0.97
24-25	0.85	0.57	0.94	0.07	0.07	0.07	0.07	0.08	1.4	13	0.31	0.91
26-27	1.1	0.63	1.2	0.18	0.17	0.19	0.18	0.18	1.6	19	0.74	1.1
28-29	0.91	0.57	1.2	0.24	0.24	0.24	0.23	0.25	1.2	12	0.31	0.85
30-31	0.65	0.57	0.95	0.09	0.10	0.10	0.10	0.11	0.79	15	0.18	0.61
32-33	0.41	0.49	0.71	0.10	0.11	0.12	0.12	0.12	0.44	9.1	0.22	0.50
34-35	0.33	0.48	0.73	0.15	0.15	0.16	0.15	0.14	0.45	9.4	0.31	0.48
36-37	0.24	0.42	0.58	0.18	0.18	0.20	0.19	0.20	0.29	6.5	0.26	0.40
38-39	0.25	0.51	0.62	1.3	1.2	1.4	1.4	1.3	0.33	7.2	0.30	0.48
40-41	0.23	0.56	0.62	0.61	0.60	0.70	0.67	0.68	0.36	7.8	0.30	0.50
42-43	0.21	0.53	0.58	0.29	0.33	0.38	0.38	0.42	0.35	7.6	0.22	0.45

Table 7 Accumulation rates of 25 elements in CNF pocosin ($\mu\text{g cm}^{-2}\text{y}^{-1}$).

Depth (cm)	Al	Ti	Mn	Fe	Mg	V	Ni	Cu	Zn	Rb	Sr	Zr	Cd
0.5-2.25	181	27	0.42	46	30	0.23	0.069	0.14	0.28	0.13	0.82	0.31	0.010
4-5	591	100	1.0	213	88	0.84	0.28	0.29	0.86	0.23	1.6	0.95	0.029
6-7	324	44	0.41	143	78	0.38	0.20	0.20	0.28	0.12	0.58	0.46	0.018
8-9	171	28	0.42	83	40	0.38	0.10	0.17	0.20	0.036	0.38	0.27	0.015
10-11	130	14	0.13	45	57	0.14	0.093	0.074	0.087	0.038	0.31	0.16	0.007
12-13	70	6.3	0.035	29	45	0.065	0.060	0.057	0.051	0.013	0.20	0.077	0.003
14-15	125	12	0.060	34	62	0.099	0.085	0.084	0.049	0.021	0.22	0.14	0.005
16-17	332	43	0.32	73	126	0.36	0.19	0.22	0.072	0.039	0.50	0.46	0.015

Depth (m)	As	Sn	Sb	La	Ce	Nd	Sm	Pr	Pb	Bi	Th	U
0.5-2.25	2.3	0.023	0.012	0.007	0.12	0.19	0.027	0.095	0.017	0.22	0.022	0.031
4-5	5.0	0.11	0.046	0.019	0.33	0.61	0.077	0.28	0.049	0.85	0.090	0.099
6-7	7.0	0.073	0.032	0.016	0.23	0.36	0.050	0.18	0.031	0.70	0.071	0.059
8-9	9.0	0.041	0.032	0.015	0.17	0.29	0.039	0.14	0.026	0.72	0.072	0.033
10-11	11	0.035	0.013	0.007	0.11	0.18	0.026	0.090	0.016	0.26	0.031	0.023
12-13	13	0.020	0.006	0.003	0.048	0.084	0.012	0.041	0.007	0.099	0.012	0.012
14-15	15	0.029	0.009	0.003	0.077	0.13	0.019	0.064	0.011	0.14	0.015	0.025
16-17	17	0.060	0.018	0.006	0.26	0.39	0.052	0.18	0.030	0.34	0.036	0.075

Table 8 Pb and Sr isotopic compositions in CNF pocosin, NC. (S.D. Standard deviation)

Depth midpoint (cm)	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{206}\text{Pb}/^{204}\text{Pb}$ S.D.	$^{206}\text{Pb}/^{207}\text{Pb}$	$^{206}\text{Pb}/^{207}\text{Pb}$ S.D.	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$ S.D.	$^{208}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$ S.D.	$^{86}\text{Sr}/^{87}\text{Sr}$	$^{86}\text{Sr}/^{87}\text{Sr}$ S.D.	Time interval
1.75	18.7961	0.0225	1.2021	0.00072	15.6446	0.0281	38.6272	0.0923	0.713475	0.000005	2001-2010
6.5	18.7423	0.0224	1.1990	0.00072	15.6324	0.0280	38.4741	0.0919	0.713168	0.000005	1976-1981
10.5	18.7606	0.0225	1.1981	0.00072	15.6584	0.0281	38.6013	0.0922	0.712338	0.000005	1942-1950
11.5	18.7606	0.0225	1.1983	0.00072	15.6565	0.0281	38.6363	0.0923	0.711959	0.000005	1938-1942
12.5	18.7366	0.0224	1.1987	0.00072	15.6302	0.0280	38.5642	0.0921	0.712036	0.000005	1930-1938
13.5	18.7627	0.0225	1.1979	0.00072	15.6624	0.0281	38.7128	0.0925	0.712100	0.000005	1922-1930
14.5	18.7347	0.0224	1.1983	0.00072	15.6346	0.0280	38.6252	0.0923	0.712270	0.000004	1915-1922
15.5	18.7521	0.0224	1.1977	0.00072	15.6564	0.0281	38.7368	0.0925	0.712461	0.000005	1908-1915
18.5	18.7619	0.0225	1.1983	0.00072	15.6575	0.0281	38.7922	0.0927	0.713078	0.000004	1883-1892
24.5	18.7369	0.0224	1.1997	0.00072	15.6180	0.0280	38.6523	0.0923	0.714035	0.000005	
28.5	18.7838	0.0225	1.1981	0.00072	15.6775	0.0281	38.8320	0.0928	0.714547	0.000006	
30.5	18.8233	0.0225	1.2020	0.00072	15.6605	0.0281	38.8153	0.0927	0.714663	0.000004	
34.5	18.9577	0.0227	1.2301	0.00074	15.6394	0.0280	39.6224	0.0946	0.716174	0.000005	
40.5	19.3915	0.0232	1.2351	0.00074	15.7007	0.0282	42.0645	0.1005	0.718209	0.000005	

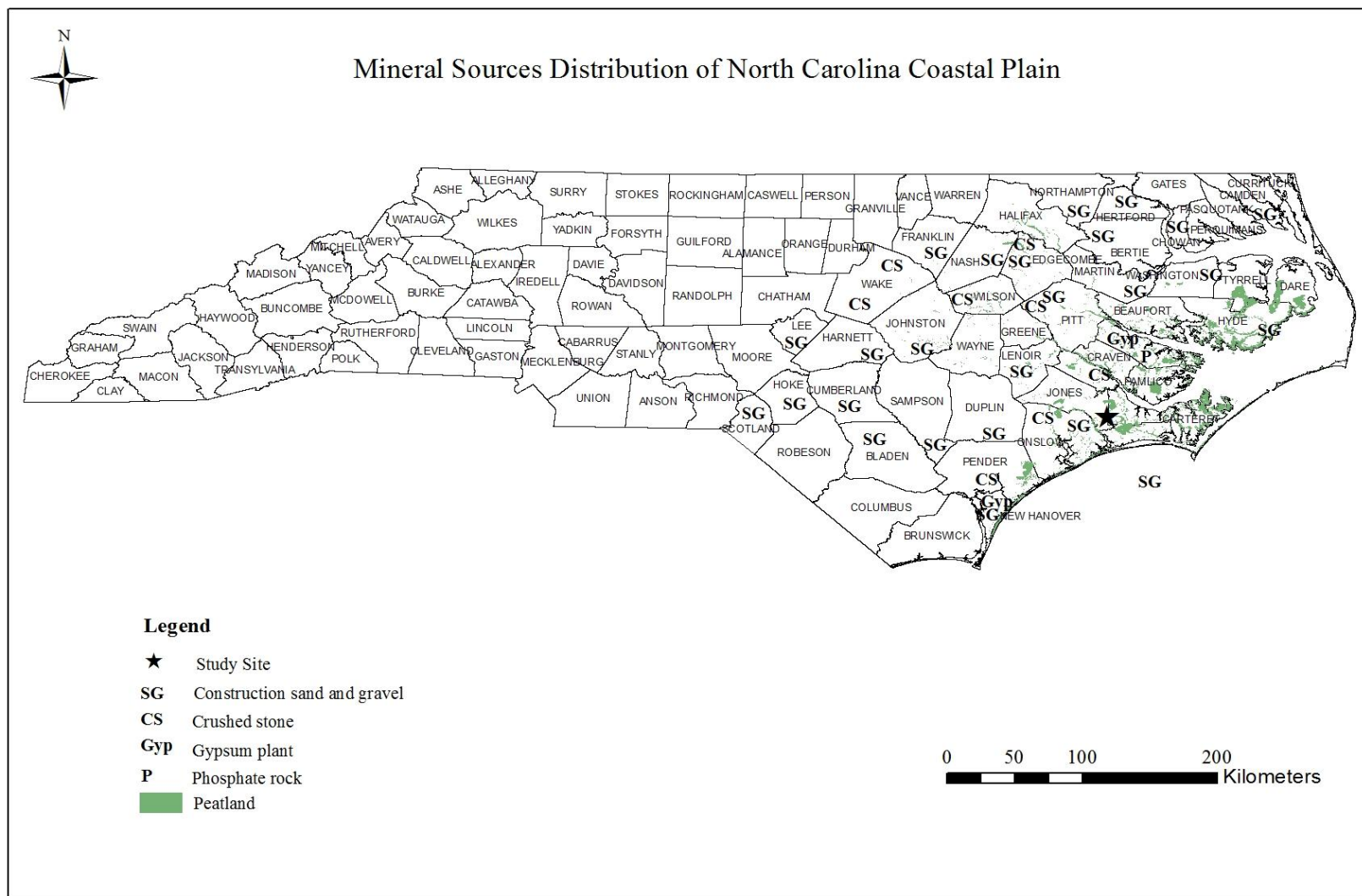


Figure 1 The mineral sources distribution of North Carolina Coastal Plain (Modification from USGS, 2012).

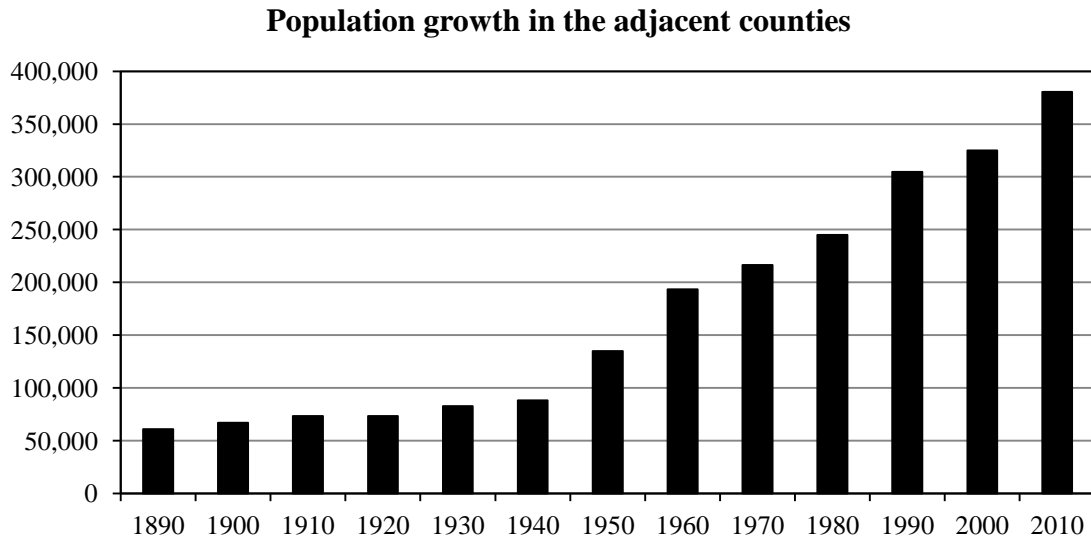


Figure 2 The total population growth in the adjacent counties around the the study site [Croatan National Forest (CNF)]: Carteret, Craven, Jones, Onslow, Pamlico.

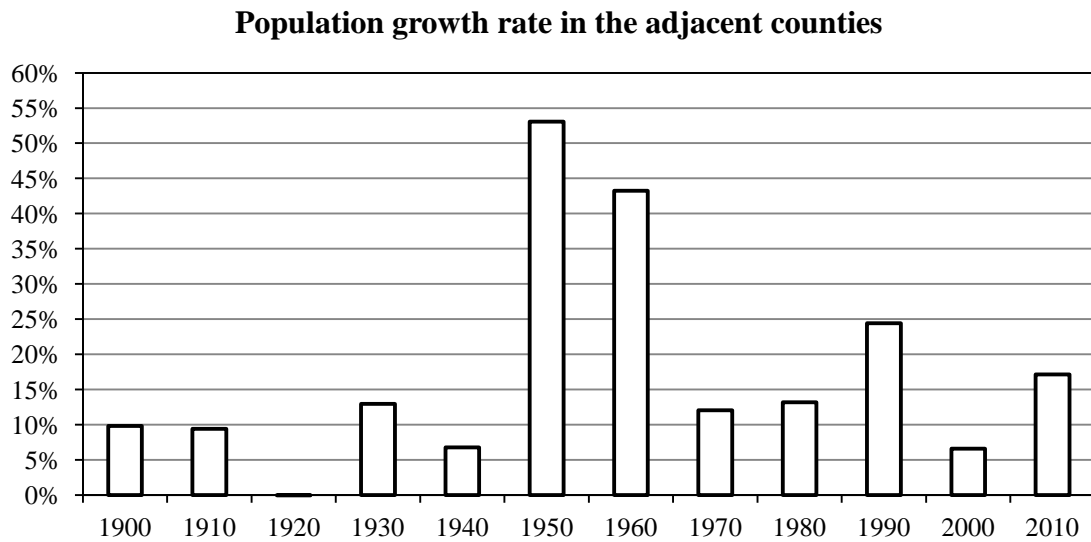


Figure 3 The total population change/growth rate in the adjacent counties around CNF: Carteret, Craven, Jones, Onslow, Pamlico.

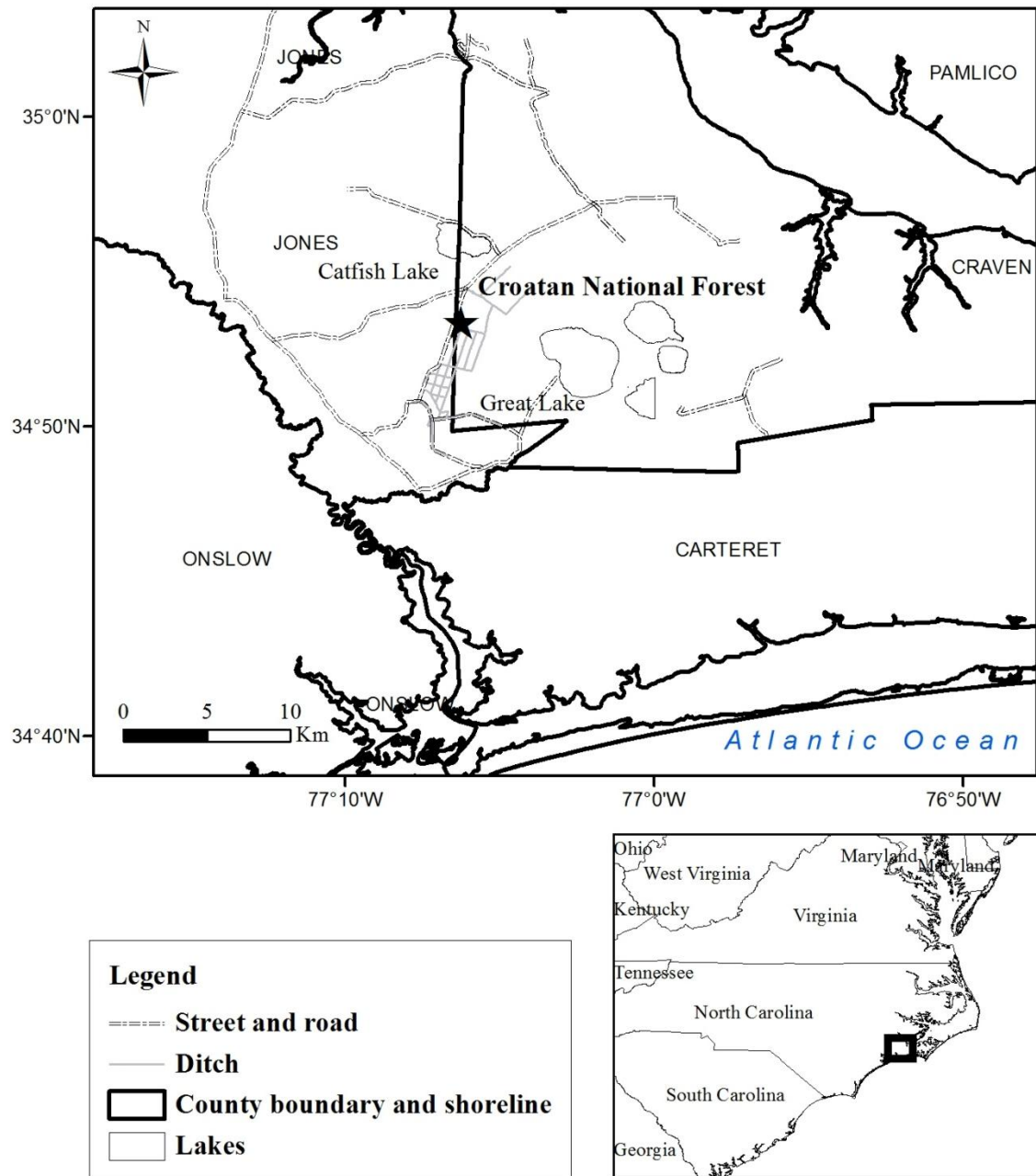


Figure 4 The study site in the pocosins of Croatan National Forest (CNF), North Carolina, USA.

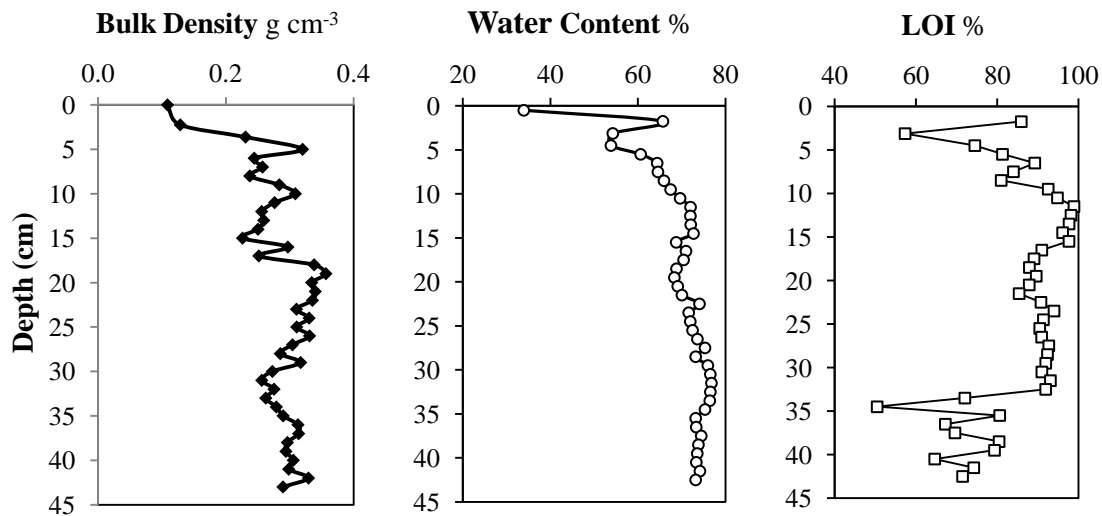


Figure 5 Bulk density, water content and Loss on Ignition (LOI) profiles of CNF pocosin.

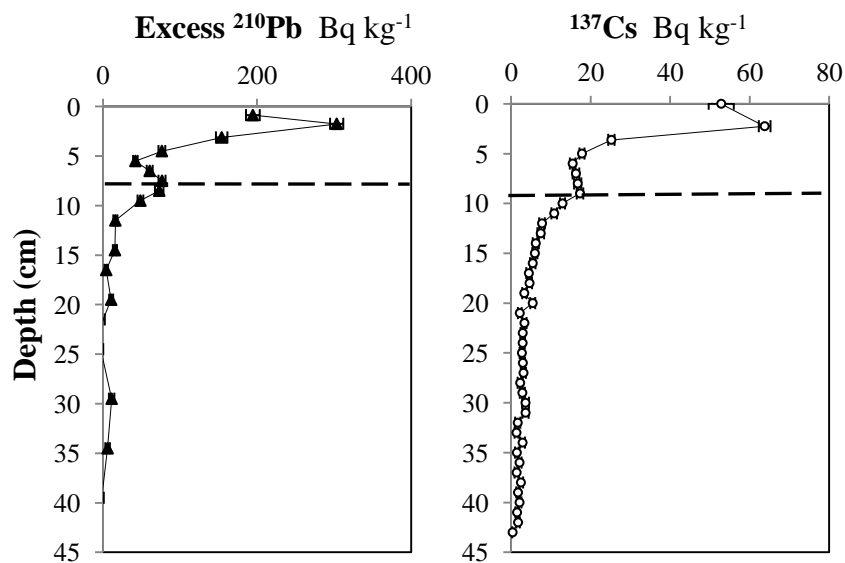


Figure 6 Excess ^{210}Pb and ^{137}Cs activities in Core 1, CNF pocosin, NC.

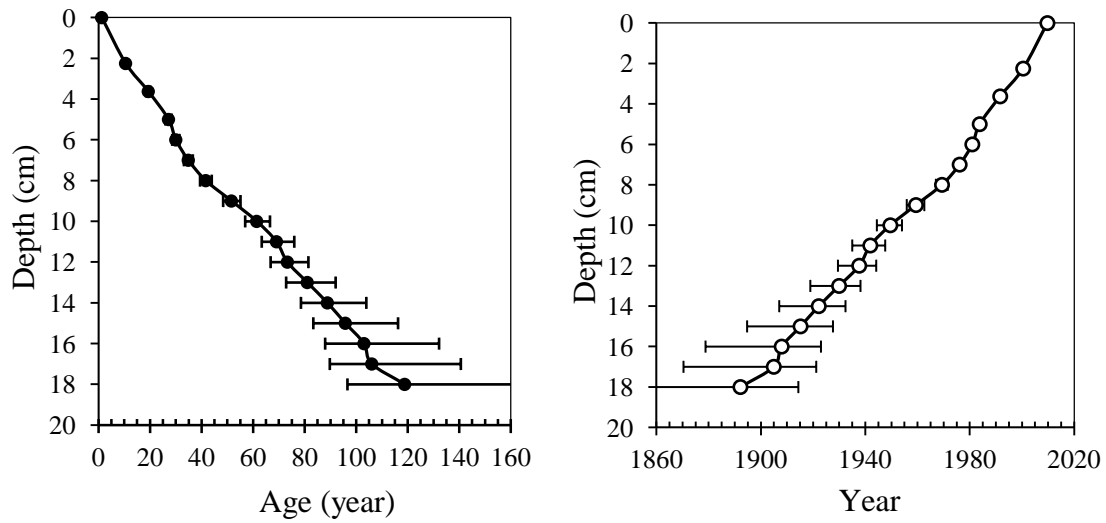


Figure 7 The chronology from CRS ^{210}Pb dating model.

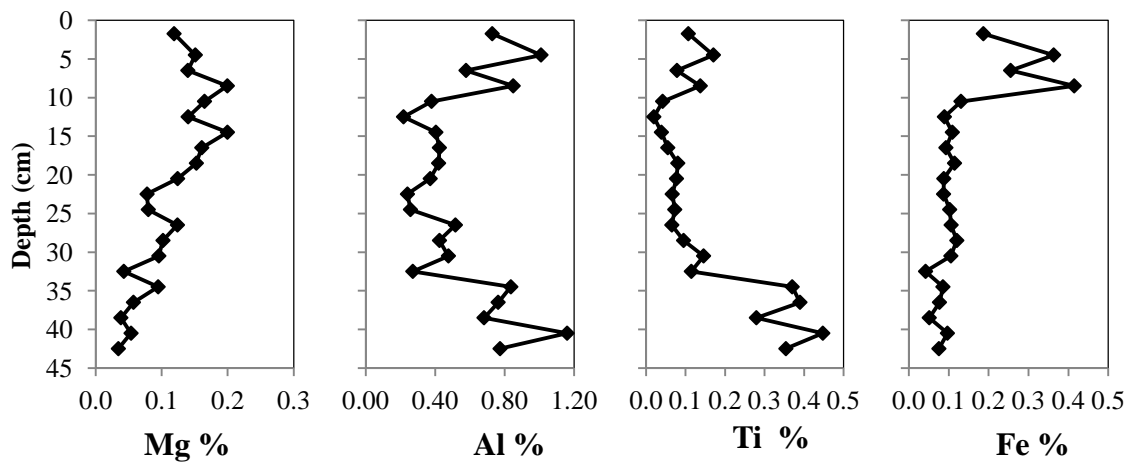


Figure 8 Major elements concentrations in peat, CNF pocosin, NC.

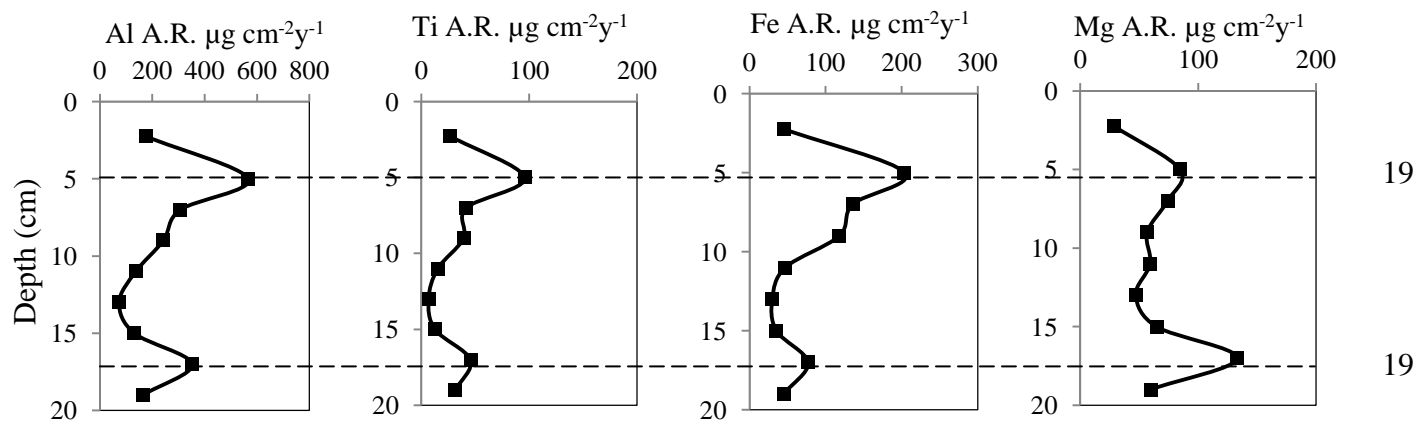


Figure 9 Major elements accumulation rates in CNF pocosin, NC. A.R. = accumulation rate

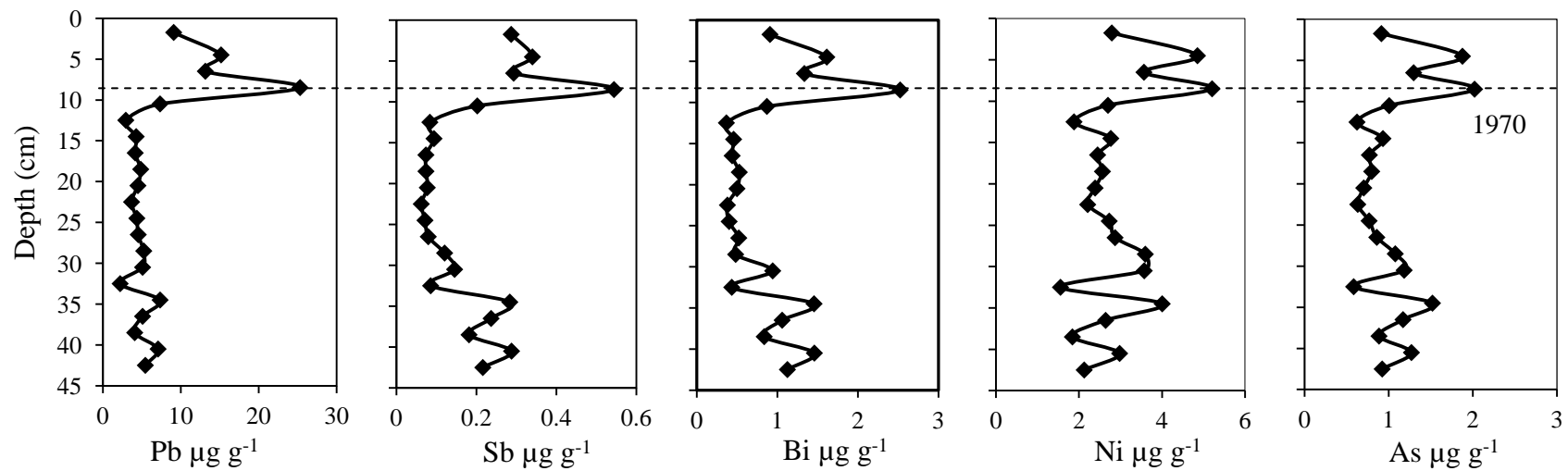


Figure 10 Selected trace elements concentrations in peat that show a significant peak at 8-9 cm, CNF pocosin, NC.

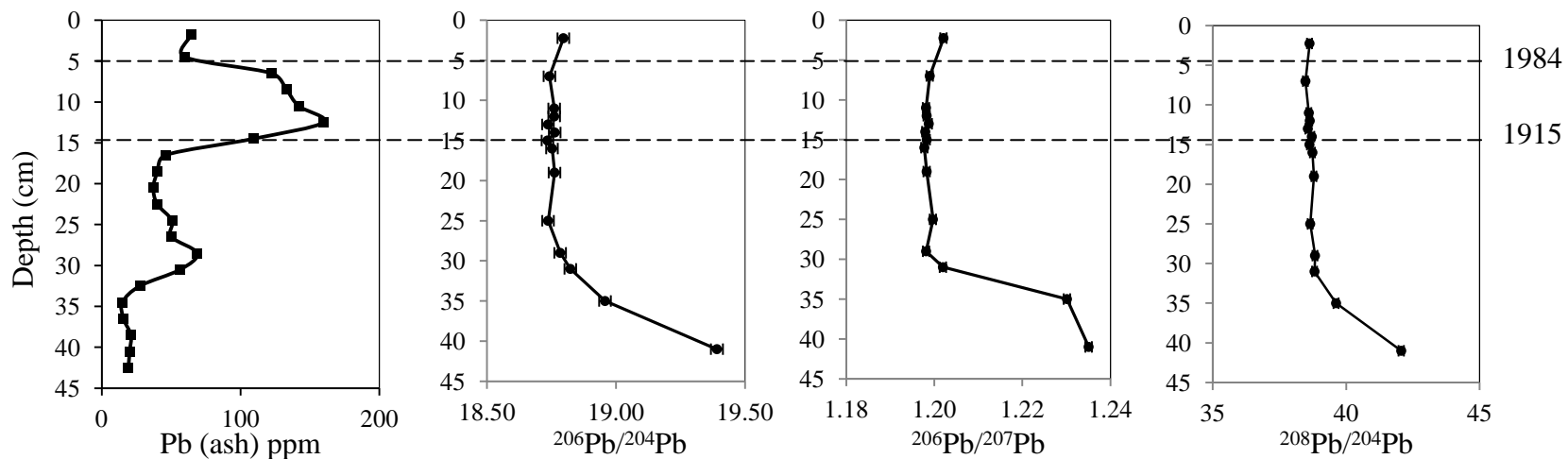


Figure 11 Pb isotopic compositions graph versus depth and age index.

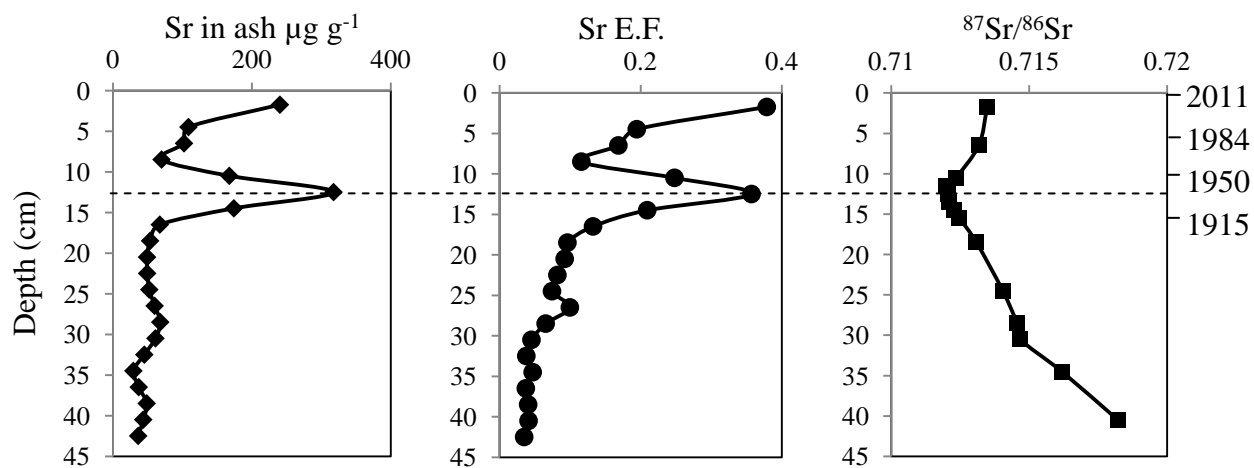


Figure 12 Sr concentration in ash, Sr enrichment factor and $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratio profiles, CNF pocosin, NC.

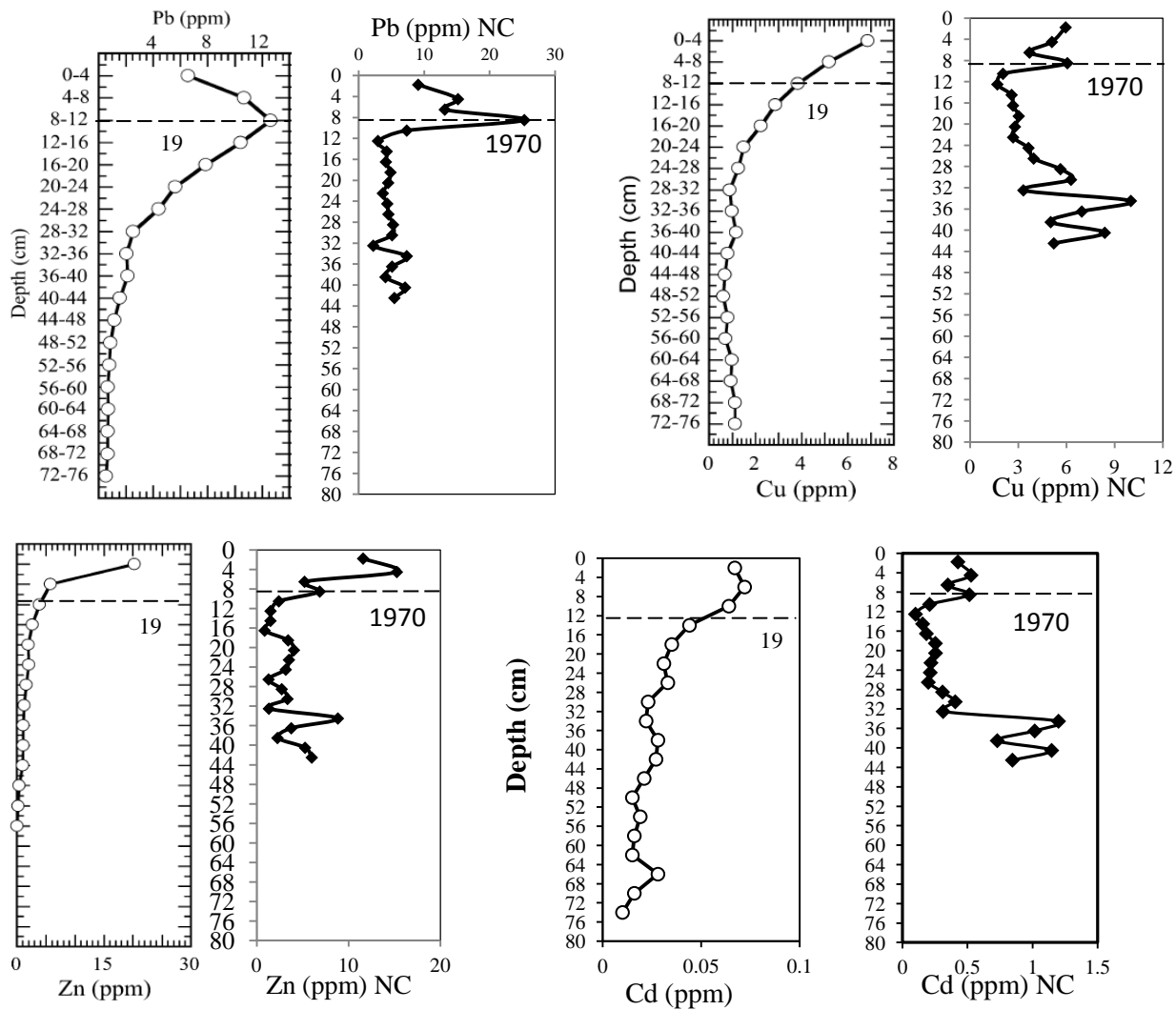
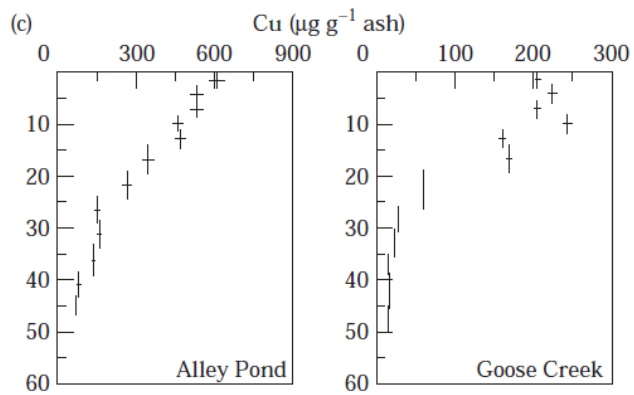
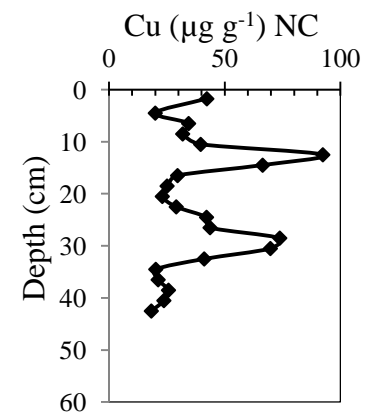
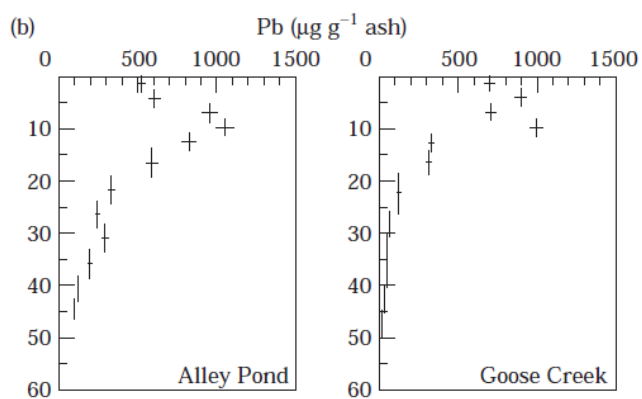
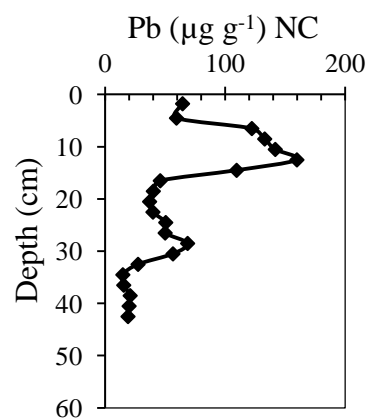
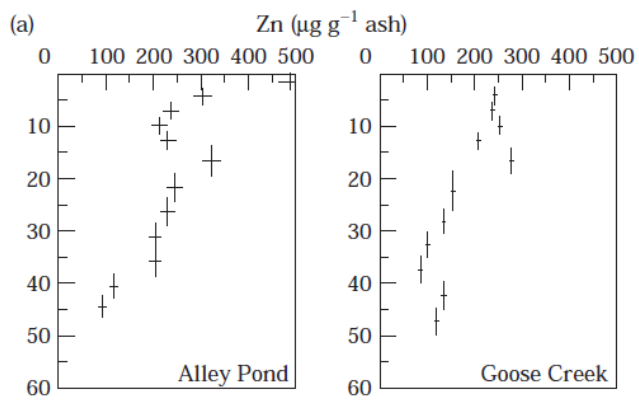
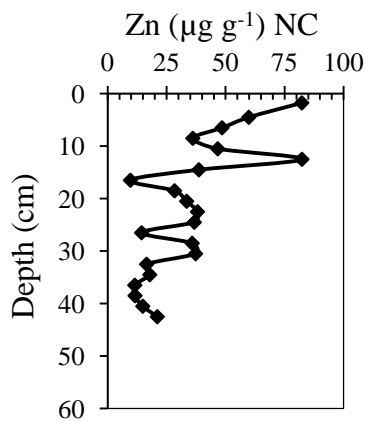


Figure 13 Pb, Cu, Zn and Cd concentrations in peat from the CNF, NC (right) graph in comparison with data from Florida (left) (Kamenov et al., 2009).



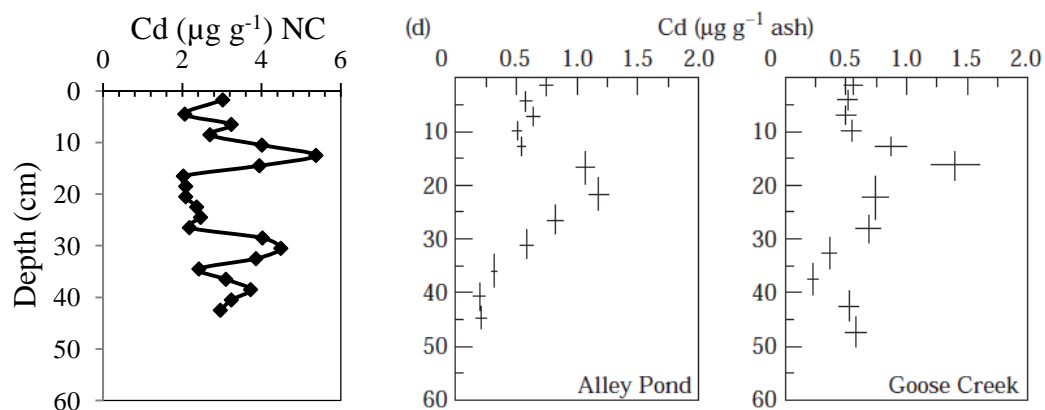


Figure 14 Zn, Pb, Cu and Cd concentrations in ash from the CNF, NC (left) graph in comparison with data from New York City (Alley Pond, Goose Creek) (Cochran et al., 1998).

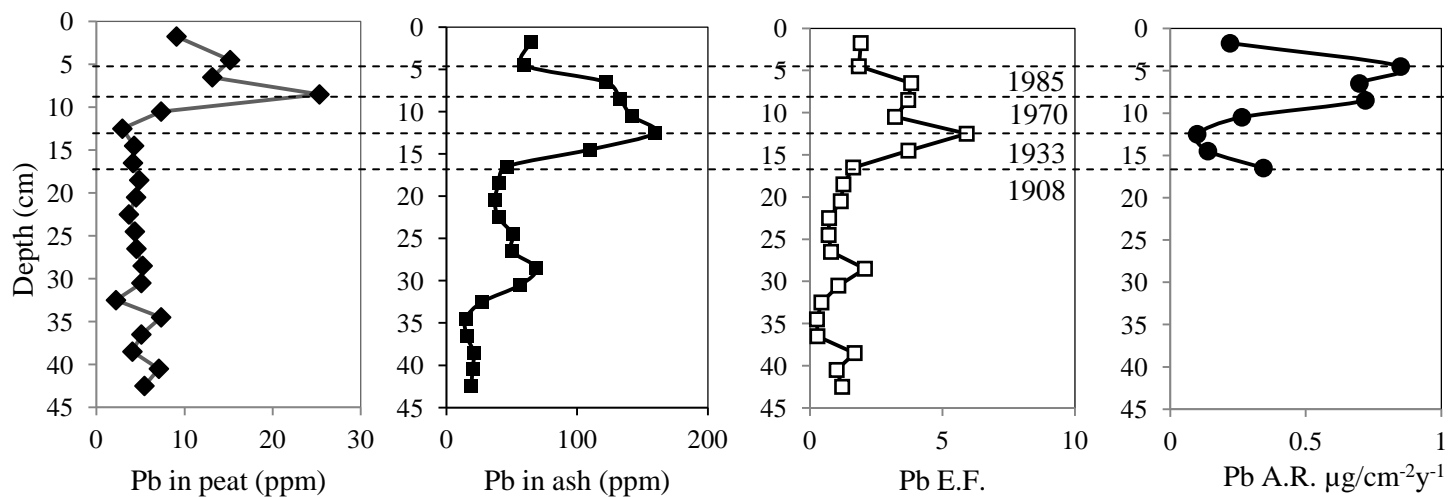


Figure 15 Pb concentration in peat, in ash, Pb E.F. (Enrichment Factor) and Pb accumulation rate in CNF pocosin, NC.

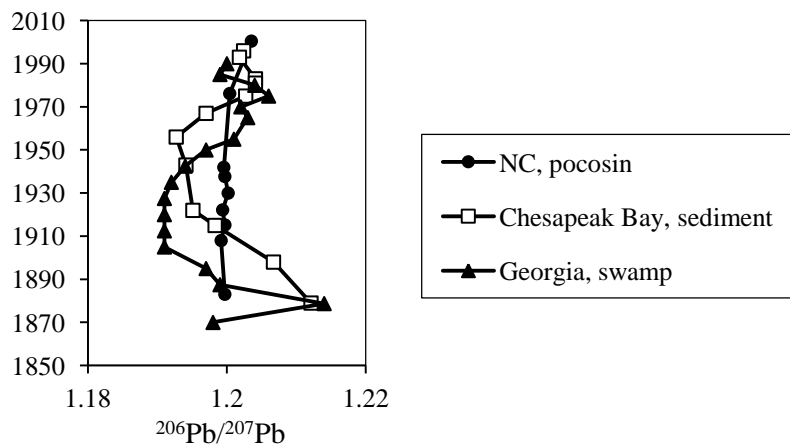
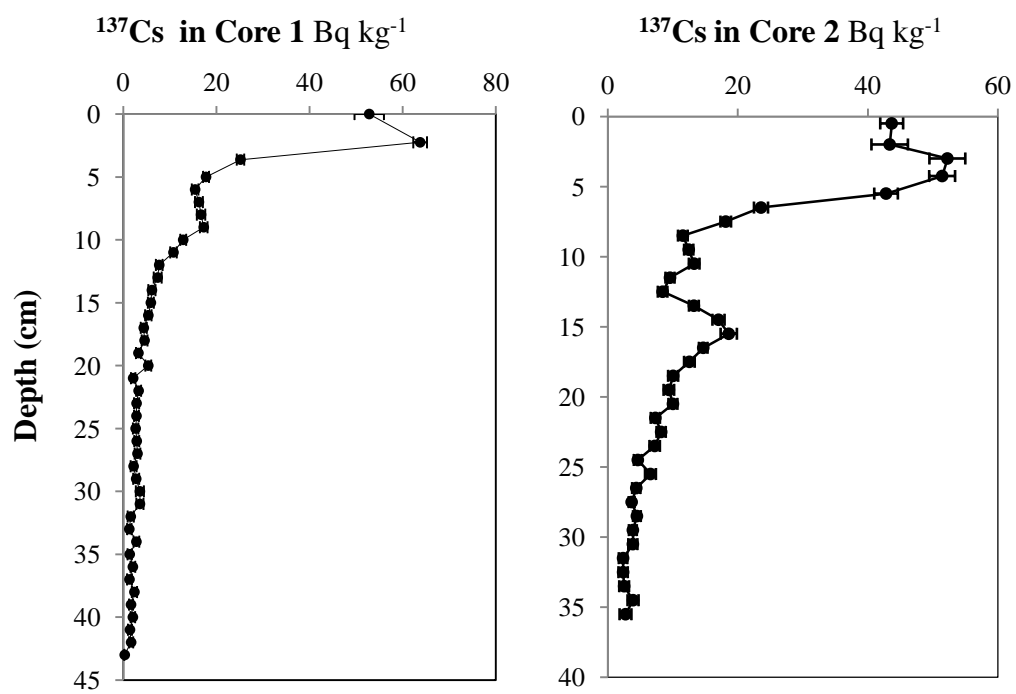


Figure 16 The $^{206}\text{Pb}/^{207}\text{Pb}$ comparisons profile between CNF NC pocosin, Georgia swamp (Jackson et al., 2004) and Chesapeake Bay sediment (Marcantonio et al., 2002).

Appendix A

^{137}Cs activities in Core 1 and Core 2 from CNF pocosin, NC.



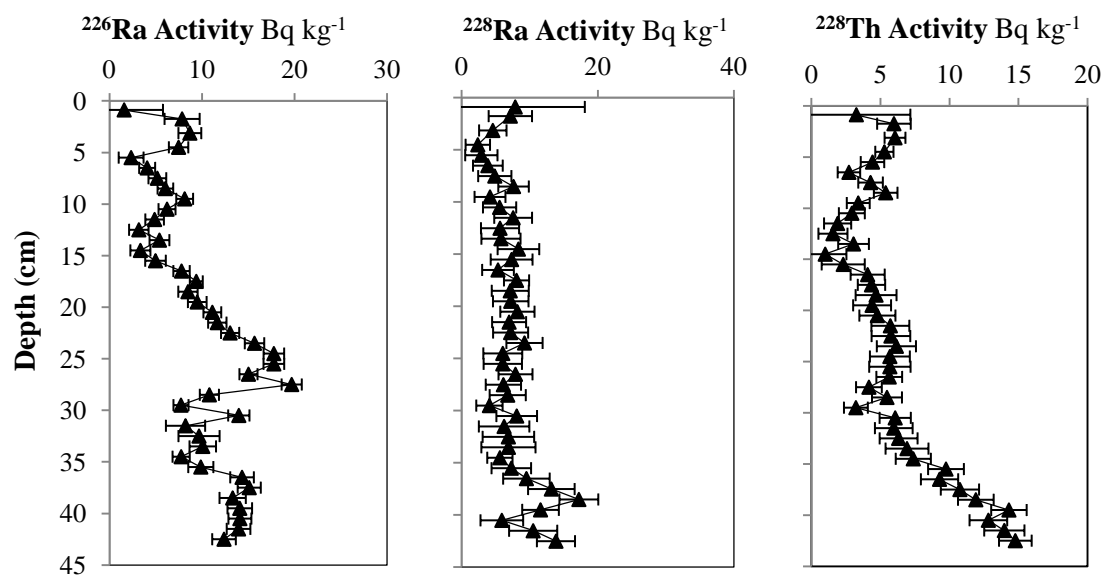
Appendix B

Table and profiles of ^{226}Ra , ^{228}Ra and ^{228}Th activities in Core 1, CNF pocosin, NC. (S.D. Standard Deviation)

Depth* cm	^{210}Pb Bq kg ⁻¹	^{210}Pb S.D.	^{137}Cs Bq kg ⁻¹	^{137}Cs S.D.	^{226}Ra Bq kg ⁻¹	^{226}Ra S.D.	^{228}Ra Bq kg ⁻¹	^{228}Ra S.D.	^{228}Th Bq kg ⁻¹	^{228}Th S.D.
0.875	194	8.91	52.8	3.16	1.59	4.20	7.88	10.2	3.25	3.91
1.75	303	8.58	63.7	1.47	7.85	1.90	7.16	3.18	5.97	1.22
3.125	154	7.45	25.2	0.796	8.68	1.23	4.59	2.02	6.05	0.756
4.5	76.4	4.42	17.8	0.640	7.47	1.05	2.38	1.79	5.28	0.660
5.5	42.0	2.92	15.4	0.706	2.34	1.34	2.90	2.37	4.42	0.851
6.5	60.5	3.87	16.2	0.845	4.07	0.864	3.86	2.18	2.71	0.815
7.5	76.5	4.57	16.7	0.898	5.17	0.961	4.88	2.44	4.28	0.891
8.5	72.9	5.53	17.3	0.816	6.04	0.845	7.63	2.24	5.37	0.856
9.5	48.5	3.74	12.9	0.630	8.12	0.909	4.17	2.27	3.39	0.830
10.5	32.2	3.18	10.8	0.702	6.22	0.918	5.58	2.44	2.92	0.940
11.5	16.0	2.62	7.73	0.714	4.88	1.00	7.57	2.78	1.89	0.984
12.5	23.8	2.92	7.40	0.849	3.17	1.05	5.65	2.80	1.55	1.05
13.5	19.6	2.79	6.15	0.766	5.40	1.07	5.78	2.87	3.05	1.11
14.5	15.4	2.66	5.93	0.637	3.34	1.08	8.35	3.06	0.993	1.54
15.5	9.77	2.45	5.38	0.687	4.97	1.11	7.35	3.07	2.30	1.56
16.5	4.12	2.24	4.40	0.648	7.79	0.883	5.35	2.33	4.07	1.24
17.5	10.1	2.51	4.58	0.622	9.38	0.700	8.07	1.85	4.35	0.992
18.5	4.88	2.54	3.26	0.527	8.48	1.05	7.15	2.72	4.68	1.48
19.5	10.5	2.57	5.38	0.630	9.48	1.01	7.22	2.61	4.39	1.37
20.5	5.26	2.43	2.13	0.469	11.1	0.962	8.20	2.52	4.77	1.31
21.5			3.30	0.528	11.7	0.993	6.98	2.50	5.71	1.37
22.5			2.86	0.545	13.0	0.982	7.20	2.57	5.75	1.40

23.5	2.84	0.523	15.7	1.05	9.24	2.66	6.15	1.42
24.5	2.66	0.583	17.8	1.11	6.05	2.86	5.68	1.45
25.5	2.90	0.528	17.8	1.12	6.05	2.82	5.68	1.50
26.5	3.06	0.610	15.0	0.977	7.92	2.50	5.63	0.933
27.5	2.23	0.636	19.7	1.09	6.14	2.59	4.16	0.933
28.5	2.78	0.636	10.8	1.03	6.77	2.65	5.47	1.09
29.5	3.57	0.865	7.73	0.813	4.09	1.94	3.22	0.865
30.5	3.58	0.762	14.0	1.15	8.10	2.99	6.06	1.13
31.5	1.61	0.661	8.22	2.12	6.24	3.71	5.96	1.37
32.5	1.30	0.480	9.68	2.22	6.88	3.78	6.31	1.37
33.5	2.83	0.606	10.1	1.43	6.88	3.98	6.92	1.55
34.5	1.39	0.462	7.74	0.929	5.63	1.86	7.38	1.27
35.5	2.06	0.508	9.87	1.36	7.30	2.91	9.74	1.30
36.5	1.34	0.523	14.3	1.28	9.52	3.42	9.28	1.35
37.5	2.40	0.562	15.1	1.25	13.2	3.42	10.8	1.39
38.5	1.67	0.433	13.3	1.42	17.2	2.85	11.9	1.29
39.5	2.06	0.480	14.1	1.33	11.6	2.69	14.3	1.29
40.5	1.44	0.427	14.1	1.24	5.91	3.13	12.8	1.36
41.5	1.69	0.442	13.9	1.28	10.5	3.54	14.0	1.46
42.5	0.317	0.101	12.4	1.28	13.9	2.80	14.8	1.18

*Midpoint depth

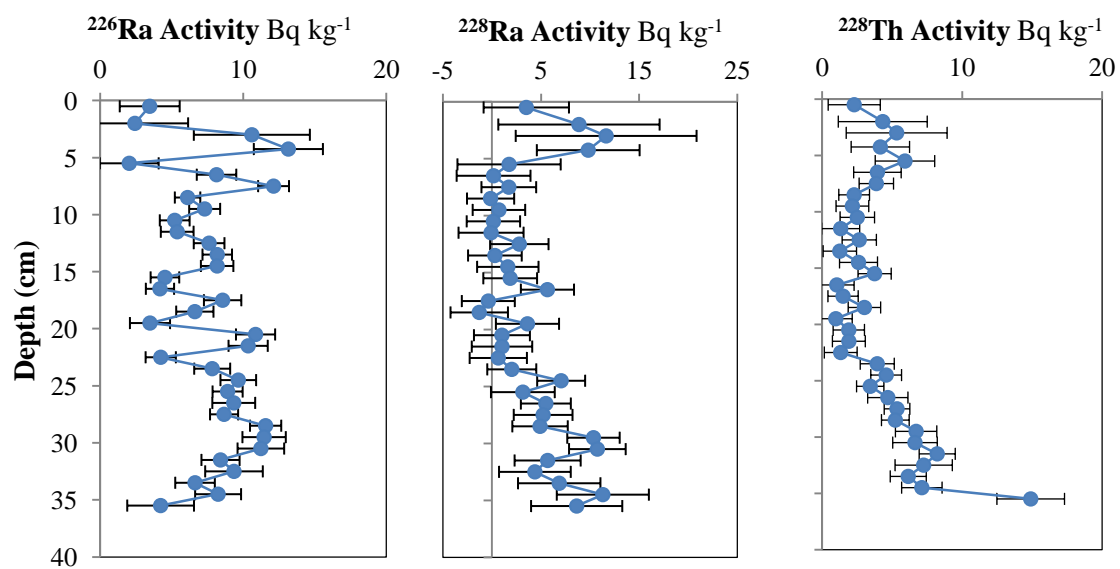


Appendix C

Table and profiles of ^{226}Ra , ^{228}Ra and ^{228}Th activities in Core 2, CNF pocosin, NC. (S.D. Standard Deviation)

Depth*	^{137}Cs	^{137}Cs	^{226}Ra	^{226}Ra	^{228}Ra	^{228}Ra	^{228}Th	^{228}Th
cm	Bq kg ⁻¹	S.D.	Bq kg ⁻¹	S.D.	Bq kg ⁻¹	S.D.	Bq kg ⁻¹	S.D.
0.5	43.7	1.76	3.46	2.10	3.50	4.35	2.29	1.86
2	43.4	2.81	2.43	3.72	8.86	8.22	4.33	3.18
3	52.3	2.76	10.6	4.05	11.6	9.21	5.32	3.61
4.25	51.5	1.99	13.1	2.41	9.81	5.24	4.16	2.09
5.5	42.8	1.82	2.03	2.06	1.75	5.26	5.92	2.13
6.5	23.6	1.08	8.13	1.38	0.168	3.76	3.95	1.70
7.5	18.2	0.813	12.1	1.08	1.72	2.79	3.87	1.23
8.5	11.6	0.742	6.11	0.890	-0.138	2.40	2.29	1.10
9.5	12.5	0.706	7.30	1.08	0.713	2.67	2.16	1.17
10.5	13.3	0.814	5.21	1.04	0.150	2.72	2.51	1.23
11.5	9.58	0.693	5.38	1.15	-0.089	3.31	1.32	1.36
12.5	8.44	0.732	7.61	1.07	2.79	2.98	2.65	1.23
13.5	13.2	0.751	8.18	1.03	0.303	2.73	1.26	1.20
14.5	17.0	0.932	8.17	1.14	1.62	3.13	2.60	1.36
15.5	18.6	1.25	4.52	1.00	1.86	2.74	3.75	1.19
16.5	14.7	0.658	4.18	0.986	5.65	2.71	1.04	1.23
17.5	12.6	0.815	8.56	1.31	-0.367	2.70	1.49	1.08
18.5	10.1	0.766	6.61	1.30	-1.293	2.92	3.02	1.16
19.5	9.40	0.807	3.48	1.40	3.62	3.22	0.974	1.17
20.5	10.0	0.687	10.9	1.37	1.02	2.85	1.90	1.12
21.5	7.32	0.702	10.3	1.37	1.03	3.08	1.91	1.18
22.5	8.21	0.705	4.23	1.06	0.647	2.92	1.32	1.17
23.5	7.22	0.797	7.83	1.26	2.02	2.50	3.94	1.22
24.5	4.62	0.602	9.65	1.25	7.05	2.44	4.58	1.10
25.5	6.57	0.833	8.90	1.05	3.15	3.25	3.44	0.978
26.5	4.38	0.631	9.33	1.50	5.49	2.55	4.69	1.44
27.5	3.67	0.448	8.66	0.981	5.22	2.99	5.35	0.917
28.5	4.48	0.591	11.6	1.09	4.90	2.82	5.23	0.989
29.5	3.86	0.601	11.5	1.51	10.3	2.67	6.72	1.48
30.5	3.86	0.698	11.2	1.63	10.7	2.89	6.64	1.59
31.5	2.35	0.650	8.41	1.33	5.67	3.37	8.22	1.29
32.5	2.36	0.711	9.35	2.01	4.38	3.65	7.25	2.05
33.5	2.52	0.750	6.63	1.38	6.85	4.20	6.15	1.29
34.5	3.89	0.847	8.24	1.61	11.3	4.69	7.12	1.44
35.5	2.73	0.910	4.22	2.33	8.63	4.64	14.9	2.42

*Midpoint depth



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