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Micro-flotation removal of coal contaminants from archaeological radiocarbon samples from Chaco Canyon, New Mexico, USA

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ABSTRACT

Micro-flotation, a specific gravity separation technique, was successfully used to remove coal contaminants from radiocarbon samples obtained from profiles, unit excavations, and solid sediment cores in Chaco Canyon, New Mexico, USA. Coal from the Cretaceous Menefee Formation occurs throughout Chaco Canyon in aeolian, alluvial, colluvial, and anthropogenic sediments. The Menefee Formation contains carbonized broadleaf angiosperm and gymnosperm plants and, as such, paleobotanical analysis was not effective in the identification and removal of coal contaminants. The effectiveness of micro-flotation as a pretreatment procedure was evaluated by: i) comparing AMS radiocarbon ages on processed and unprocessed samples from the same archaeological contexts; ii) comparing a processed sample of carbonized hardwood charcoal with a sample of uncarbonized hardwood from the same archaeological context; and iii) comparing radiocarbon ages on a split sample of processed bulk carbon. The comparisons confirmed the effectiveness of micro-flotation in processing samples for radiocarbon dating in Chaco Canyon and would be applicable for similar locations elsewhere in the world.

1. Introduction

Coal occurs in sedimentary and occasionally in low-grade metamorphic rocks. Coals are composed of macerals (inertinite, liptinite, vitrinite), which are predominantly plant matter, tissues, spores, pollen, resins, and humus that were inundated, buried and compressed (Thomas, 2012). In time, macerals are carbonized with increasing pressure, temperature, and acidity. As these processes increase, the plant remains are transformed into different grades of coal—progressively peat, lignite (also known as brown coal), sub-bituminous, bituminous, anthracite, and graphite (Stach et al., 1975).

Coal deposits occur around the world and date from the Proterozoic (~2 Ga) to the Pliocene (~2 Ma) (Tyler et al., 1957). Consequently, the original atmospheric radioisotope ^{14}C is absent in coal since its half-life is ~5730 years. While trace amounts of ^{14}C occur in coal, they are attributed to cluster decay from radiogenic isotopes, which create ^{14}C from ^{12}C (Beck, 2011). Additionally, bacteria (*Diplococcus* sp.) and fungus (*Trametes versicolor*) metabolize and degrade coal (Potter, 1908), which can artificially enrich the ^{14}C content of coal (Campbell et al., 1988).

Chemically, coal is composed of C, H, N, S, O, and about 140 different hyrdrocarbons such as anthrocene, benzene, ethylbenzene, n-hexane, 2-hexene, methyl ethylbenzene, naphthalene, propylbenzene, and toluene (Tankersley and Munson, 1992). The quantities of these chemicals vary from one grade of coal to the next and even from one lump of coal to the next (Tankersley and Munson, 1992). Isotopically, the composition of coal includes ^{12}C, ^{13}C, and trace amounts of post-depositional ^{14}C.

Radiocarbon ages are calculated by determining the ratio of ^{12}C to ^{14}C in an archaeological or geological sample. Consequently, a natural or anthropogenic admixture of coal would likely increase the quantity of ^{12}C and result in an age determination that was older than expected (Tankersley et al., 1987). The geologic distribution of coal is pan-global and poses a significant contamination threat to accurate radiocarbon

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Table 1
Specific gravity of wood charcoal and different grades of coal.

Sample	Specific gravity[a]
Wood charcoal	0.40
Peat	1.24
Lignite (Jet)	1.29
Sub-bituminous coal	1.30
Bituminous coal	1.32
Anthracite	1.47
Graphite	2.25

[a] Averages after Wood et al. (1983).

dating in many regions. Coal can occur in samples collected for radiocarbon dating as the result of weathering, erosion, deposition processes, groundwater transport, and human introduction through its procurement as a fuel source or a manufacturing raw material (Tankersley, 1984; Tankersley et al., 1987).

Archaeologically, the oldest use of coal is uncertain. Jet was used in carvings in the Shenyang region of China ~4000 BCE and as a fuel source by ~1000 BCE (Golas and Needham, 1999). In eastern North America, bituminous coal was used to manufacture beads as early as the Late Archaic cultural period (e.g., the DuPont site, 33Ha11, ~3000 BCE; Dalby, 2007). By ~1000 CE, coal, coal ash, and "clinkers" occur in hearth features of Late Woodland and subsequent Fort Ancient and Mississippian sites suggesting that it was used as a fuel source (Tankersley et al., 1987).

Coal can be indistinguishable from more recent carbonized plant remains in archaeological and geological radiocarbon samples, even under high magnification scanning electron microscopy, because it is composed of carbonized plant matter. While prolonged acid-base-acid sample pretreatment can eliminate contamination from coal humates, it is an ineffective procedure for the removal of particulate and larger coal particles (Tankersley et al., 1987). Paleobotanical analysis can be used to successfully identify distinctive residues of fossil algae and spores if radiocarbon sample contaminants are from Proterozoic and Paleozoic coals, respectively (Tankersley et al., 1987; Tankersley and Munson, 1992). However, contamination of radiocarbon samples from Cretaceous and Tertiary coals pose a greater challenge because of the presence of angiosperm and conifer plant fossils that may overlap with archaeological plant assemblages. In these situations, density or specific gravity—the ratio of the mass of a material to the mass of a reference material for the same given volume—can be used to separate coal from particles of more recent carbonized plant remains of a differing specific gravity (Table 1).

Flotation is the most common specific gravity separation technique used by archaeologists and geologists to recover botanical remains. Samples are placed in water, which is gently circulated, separating carbonized plant remains from sediments with a higher specific gravity onto a filter by using a fine mesh screen. Radiocarbon sample sizes (≥50 g for conventional β-decay and ≥5 mg for accelerator mass spectrometry [AMS]) are far less than those obtained for archaeobotanical and paleobotanical analyses so flotation separation systems need to be greatly scaled down to what we call micro-flotation to recover all possible usable samples.

2. Methods

2.1. Chaco Canyon

We collected a suite of radiocarbon samples from Chaco Canyon, located in the central San Juan Basin of northwest New Mexico, to determine whether micro-flotation can be successfully used to separate archaeological and geological carbonized plant remains from coal contaminants (Fig. 1). Chaco Canyon is formed in the Cretaceous Cliffhouse

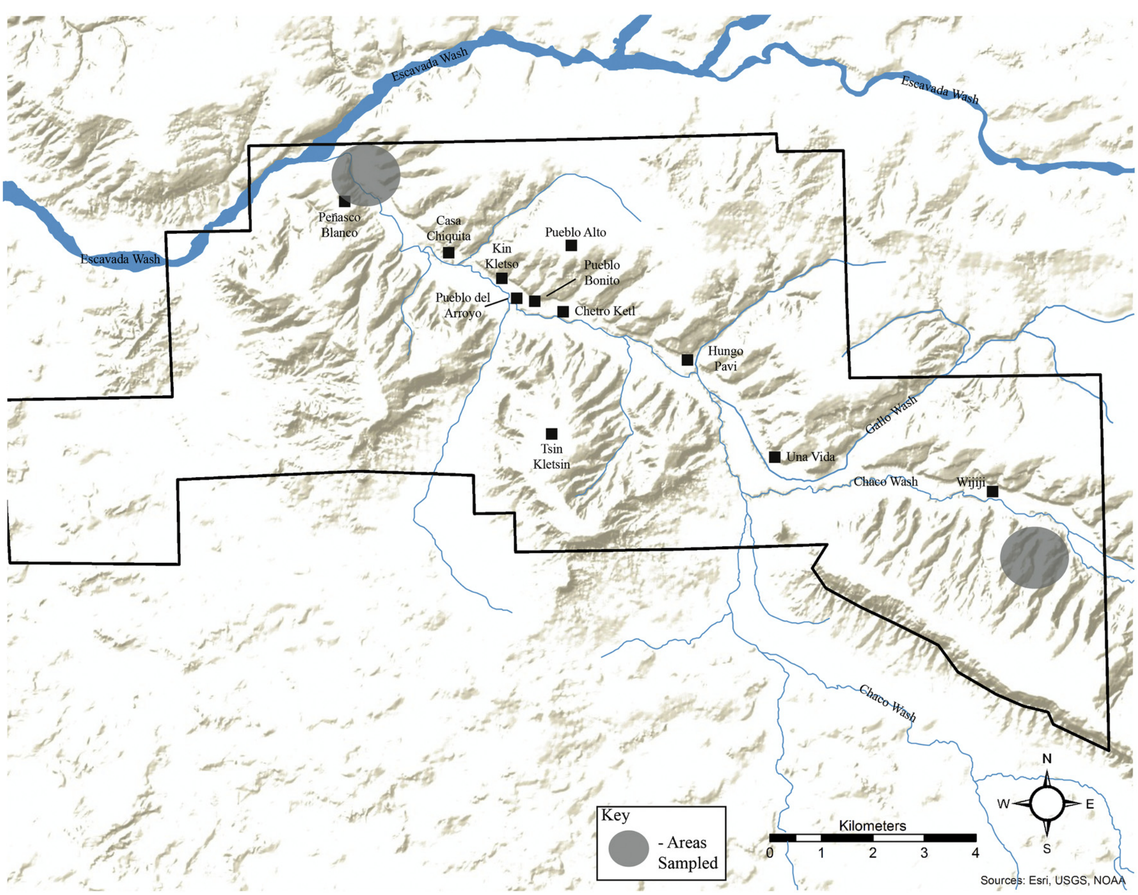

Fig. 1. Geographic location of the study area.