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A preliminary assessment on the use of biochar as a soil additive for reducing soil-to-plant uptake of cesium isotopes in radioactively contaminated environments

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Abstract A series of K_d tracer batch experiments were conducted to assess the absorptive-desorption properties of Biochar as a potential agent to selectively sequester labile soil Cs or otherwise help reduce the uptake of Cs isotopes into plants. A parallel experiment was conducted for strontium. Fine-grained fractionated Woodlands tree Biochar was found to have a relatively high affinity for Cs ions ($K_d > 100$) relative to coral soil ($K_d < 10$) collected from the Marshall Islands. The Biochar material also contains an abundance of K (and Mg). These findings support a hypothesis that the addition of Biochar as a soil amendment may provide a simple yet effective method for reducing soil-to-plant transfer of Cs isotopes in contaminated environments.

Keywords Marshall Islands · Biochar · Remediation · ^{137}Cs · Cesium distribution coefficients · K_d Values

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Introduction

Pyrogenic carbon (Cpyro), commonly known as Biochar or Terra Mulata, is a porous charcoal that has been derived from partial carbonization of ligno-cellulosic materials including plant and waste biomass through pyrolysis. The use of Biochar in agriculture dates back to between 450 BC and 950 AD forming part of the Ancient Amazonian Dark Earth soils distribution (Amazon Black Earth) [1–3]. The humic components derived from Black Carbon have high aromaticity and charge density giving rise to high CEC values and sustainable fertility. Biochar is extremely stable in soil and is known to greatly improve the productivity of plants. There is also a growing interest in Biochar research as a carbon-negative pathway for semi-permanent storage of atmospheric CO_2 [4]. Moreover, a number of previous studies have shown that Biochar has a high capacity to absorb heavy metal and organic pollutants from soil [5, 6]. In this study, we have explored the remedial properties of Biochar as a soil amendment to potentially help reduce the soil-to-plant uptake of ^{137}Cs contained in aged fallout deposition on coral atolls in the Marshall Islands.

Coral atoll soils are largely composed of Ca–Mg–Sr carbonates with variable quantities of organic matter. The Al–Si based clay mineral content of coralline atolls is also not easily recognizable. Most coral soils are deficient in K and generally lack a number of other essential trace elements and micronutrients to support sustainable cultivation of food plants [7, 8]. Horticultural practices are also constrained by the low water holding capacity of sandy coral soil. The deficiency in K and absence of clay minerals yield conditions that enhance the uptake of ^{137}Cs into plants similar to that found in peat and other rich organic soils [9, 10]. Transfer factor (TF) values of ^{137}Cs from soil-to-plant for most tree-crop foods such as coconut, breadfruit and *Pandanus* fruit in the Marshall Islands typically range between 1 and 40 [11]. Measured ^{137}Cs TF values for vegetables and grains are more variable and have been known to exceed 100 [12, 13]. By comparison, ^{137}Cs TF values for similar types of plants growing in continental mineral soils typically range from about 0.004 to 0.5 [12, 14] or several orders of magnitude less that those observed in the Marshall Islands. Subsequently, the main controlling factors in assessing the dose contribution to resettled and resettling populations in the northern Marshall Islands are diet and the uptake of ^{137}Cs into locally grown foods [15–19]. External gamma exposure to residual ^{137}Cs contamination in soil is less important, accounting for about 5–15 % of the total nuclear test-related dose [15, 20].

The United States conducted 67 atmospheric nuclear tests at Bikini and Enewetak Atoll in the northern Marshall Islands [21]. Under the auspices of the Office of Health and Safety (AU-10) at the U.S. Department of Energy (DOE), Lawrence Livermore National Laboratory (LLNL) scientists continue to provide radiological protection monitoring of local inhabitants and the environment in the Marshall Islands [16–18]. Key program research directives are to build a strong technical and scientific foundation to help support safe and sustainable resettlement programs, and improve food safety and security [8, 15, 19]. The treatment of agricultural areas with K-rich chemical fertilizers has been promoted as the most effective method to reduce the uptake of ^{137}Cs in tree food crops such as coconut, breadfruit and *Pandanus* fruit [15, 22, 23]. Motivation for the present study was prompted by the need to reduce the uptake of soil ^{137}Cs into cultivated plants such as garden vegetables without the need for the addition of large quantities of chemical fertilizers, and to help conserve irrigation water.

Experimental

A series of radioactive tracer experiments using ^{134}Cs and ^{85}Sr were designed to determine the distribution or partitioning coefficients (commonly referred to as a K_d in pelagic environments) of Cs^{+} and Sr^{2+} ions on different size fractions of Biochar and coral soil suspended in Bikini soil water. The Biochar material was supplied by Mr. Daniel Hawk, Space and Earth Carbon Research Environmental Team, Oneida, WI (Oneida Indian Reservation) (Fig. 1). The material was derived from U.S. Midwest woodland trees also known as Northwoods—mostly oak, red oak and other shade-tolerant and woody native species. The soil samples used in this experiment were collected from the interior of Bikini Island. The soil was air dried and homogenized in a V-mixer prior to use. Separate sub-

Fig. 1 Biochar material produced by Dan Hawk, New Dark Earth, Space and Earth Carbon Research Environmental Team, Oneida, WI 54155, USA (Oneida Indian Reservation)

aliquots of Biochar were taken for preparing scanning electron microscopy (SEM) micrographs, and for measuring various chemical and physical properties of the Biochar. The SEM system consisted of an Hitachi SU-70 scanning electronic microscope containing a thermal field source and a back scattered electron (BSE) detector. Potassium (K), calcium (Ca), magnesium (Mg), copper (Cu), zinc (Zn), manganese (Mn) and iron (Fe) were determined by atomic absorption spectroscopy. Phosphorus (P) was analyzed colorimetrically using a Technicon II CFI AutoAnalyzer. Total N was determined by the Kjeldahl method. Electrical conductivity (EC) was measured using a Walden precision apparatus (WPA) meter and probe. The percent by weight of organic matter was determined by Walkley–Black or Loss on Ignition. Brunauer–Emmett–Teller (BET) surface area measurements were performed on pulverized material that passed through a 63 µm sieve. The Bikini soil water (salinity ~0.1 ppt) was collected from an underground catchment basin connected to large-size plate lysimeter inserted into the soil at a depth of about 1 m. Separate plant growth experiments were conducted to determine the influence of Biochar on plant growth, seed germination, vegetative growth, fungal associations (e.g., *Arbuscular mycorrihizal)* and on soil-to-plant uptake of ^{137}Cs (to be published elsewhere).

The Biochar was size-fractionated by dry sieving for 20 min using an automated Sieve Shaker fitted with >4.75, 1.18, 500, 320, 125 and 64 µm size standard mesh screens. A bulk sample of biochar was also prepared by passing pulverized material through a 64 µm sieve screen. Tracer solutions of ^{134}Cs and ^{85}Sr were prepared by serial dilution from known concentrations of the stock solutions supplied by Eckert & Ziegler Isotope Products (Valencia, CA). The nominal spike addition used for all distribution coefficient experiments was 600 Bq each of ^{134}Cs and ^{85}Sr tracer. Our standardized protocol called for Biochar and soil samples to be wetted and equilibrated before use by shaking with Milli-Q water (1:30 ratio) for 24–48 h. The material was then separated by centrifugation and the wetting procedure repeated for a second time.

The tracer distribution experiments were carried out in 250 mL Erlenmeyer flasks sitting on a rotating table. Each experiment was conducted with 5 g of Biochar and 100 mL of 0.2 μm filtered soil water adjusted to pH 7.0 after addition of tracer solutions. Distribution coefficients were first measured after adding the Biochar to the aqueous phase and gently mixing the sample by hand (time = 0) followed by measurements performed at internals over the next 1700 h (~70 days). Each sample consisted of 1–1.2 mL of solution drawn up from the sample flask using a plastic pipette, and then 0.2 μm syringe filtered into pre-weighed, standard geometry counting vials. The vials were then made up to volume using 2 % ultrapure HNO_3 and counted by high-resolution gamma-spectrometry to determine the total amount of ^{134}Cs and ^{85}Sr present in the aqueous phase. The quantity of tracer absorbed onto the Biochar was then back-calculated using a total activity mass balance equation, and the distribution coefficient reported as Bq g^{-1} in Biochar divided by Bq mL^{-1} in water (expressed as a K_d, in units of mL g^{-1}). After termination of the absorption experiment, the remaining Biochar was filtered, air dried under vacuum and then re-suspended in fresh soil water with no added tracer. A desorption experiment between the Biochar and soil water was then conducted in a similar fashion to that described above.

Results

Particle size data show 75 % of the stock Biochar being retained on a 4.76 mm sieve with only about 5 % being less than 500 μm. On a dry weight basis, the Biochar contained about 70.6 % organic matter ($C/N = 75.5$) and had bulk dry density of about 207 kg m^{-3}. The surface area of pulverized Biochar material passing through a 63 μm sieve averaged about 193 m^2 g^{-1}. Surface area was shown to increase with decreasing particle size. For example, the measured conductivity (EC) for Biochar material around 2 mm is about 124 μS cm^{-1}, increasing to 474 μS cm^{-1} for material passing through a 64 μm sieve. A typical SEM micrograph of Northwoods Biochar is shown in Fig. 2. The Biochar contained 36 ppm total N, 71 ppm total P, 1670 ppm of K (2021 ppm K_2O), 613 ppm of Ca, 106 ppm of Mg, 0.5 ppm of Cu, 3 ppm of Zn, 34 ppm of Mn, and 9 ppm of Fe.

The results of the ^{134}Cs (and ^{85}Sr) distribution coefficient experiments performed on two different size fractions

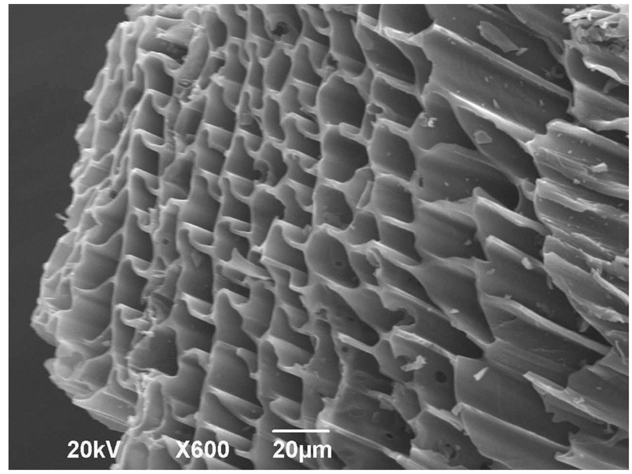

Fig. 2 SEM of Northwoods tree biochar (Carbolea Group, Ireland)

and a pulverized sample of bulk Biochar, and two surface soil sample collected from Bikini Island, are shown in Fig. 3. Results from the ^{85}Sr partitioning experiment are only shown for the <120 um size fraction Biochar and a single soil sample.

Discussion

Biochar is composed of polyaromatic units of different size and with different organizational levels [24]. SEM photos show that the Biochar used in this tracer experiment has excellent porosity. The network structure of Biochar provides a very large surface area for surface site reactivity. The pores of Biochar also provide a refuge for water and *Arbuscular mycoorhizal* fungi in association with plant roots—effectively extending the root and helping enhance the uptake of nutrients to the plant. The addition of Biochar is likely to greatly enhance the water holding capacity of sandy coral soil. However, in this instance, the available nutrient content of the Biochar is very low except for an abundance of K and Mn. Nonetheless, Biochar added as soil amendment to coral soil may provide adequate K to help suppress soil-to-plant uptake of ^{137}Cs and support healthier plant growth. The effectiveness of added K on reducing the uptake of ^{137}Cs in locally grown tree-crop foods in the northern Marshall Islands has been previously demonstrated in large-scale remediation experiments [22, 23].

Distribution coefficients of Cs in soil and sediments in the natural environment may vary over several orders of magnitude [25–27]. The experimental values measured for Northwoods tree Biochar tend to be aligned with the lower range of values reported. Cs absorption on soil (and sediment) may be affected by many different abiotic and biotic processes including physical and/or biogeochemical such

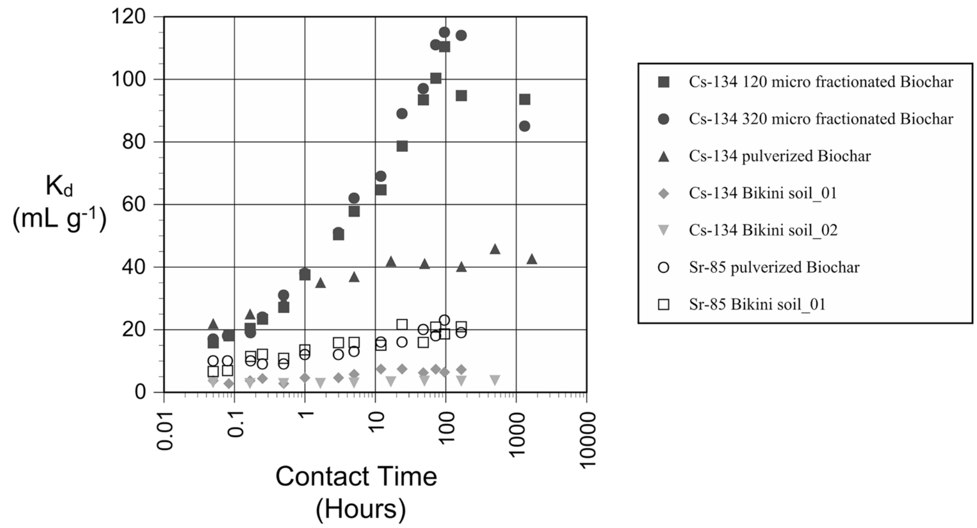

Fig. 3 Distribution coefficient (K_d) measurements of ^{134}Cs and ^{85}Sr on different forms of Northwoods tree biochar and Bikini soil

as soil mineralogy and organic content, grain-size, cation-exchange capacity, pH and the presence of coexisting cations such as K, Na, Mg and Ca [28–31]. Important soil constituents affecting the sorption of Cs include the presence of minerals such as smectite, illite and chlorite [32–34]. The sorption characteristics of radionuclides under experimental conditions may also be affected by the soil/solution ratio, contact time, and the initial tracer or stable analog concentration [31, 35–37].

The distribution coefficient experiments using fine screen size fractionated Biochar clearly show enhanced absorption of ^{134}Cs on Biochar compared with that observed with untreated Bikini soil. The measured distribution coefficient (expressed as a K_d value) for ^{134}Cs after a 7-days contact time approached 1×10^2 mL g^{-1} or a factor of 10–20 times higher than that observed in soil samples ($K_d < 10$). The short-term equilibrium K_d for the bulk pulverized Biochar was about half the value observed for fractionated Biochar. These later results may be more reflective of the Cs sorption characteristics of the bulk Biochar. It is also of interest to note that greater than 95 % of the sequestered ^{134}Cs on Biochar was retained during desorption experiments. Moreover, the amount of ^{134}Cs lost to solution during desorption experiments could easily be accounted for by washout of interstitial fluids containing unabsorbed tracer solution from the original K_d experiment. This shows that the mechanism for Cs ion absorption is largely irreversible over the timescale of the experiment. The lower ^{134}Cs K_d values observed for fractionated Biochar after contact times in excess 100 h appear to be related to changing pH conditions. An increase in soil pH over time was also observed in associated greenhouse plant growth experiments (Hayes—personal comm.).

Maximal K_d values measured for ^{85}Sr on pulverized Northwoods Biochar were about half those observed for ^{134}Cs, and were indistinguishable to those measured for coral soil (Fig. 3). Stable Sr is a major chemical constituent of coralline soil (and coralline soil water). The lower K_d values observed for ^{85}Sr relative to ^{134}Cs on Biochar can be explained by differences in the absorption properties of the elements or ions, on the selectivity and relative number of available absorption sites for each element or ion, and on mass-action effects or presence of stable Sr in competing for available absorption sites on the Biochar.

Soil-to-plant transfer factor (TF) values for ^{137}Cs in the Marshall Islands are at least 1–2 orders of magnitude higher than that observed for the similar plants growing in mineral soils from continental regions [11–13]. The difference in behavior of ^{137}Cs can be attributed to the unique nature of coral soils in maintaining an available pool of labile ^{137}Cs for soil-to-plant uptake. Plants transport labile ^{137}Cs across root membranes as a natural chemical analog to K. Coral soils are naturally deficient in K so there is less chemical competition for soil-to-plant uptake of ^{137}Cs in satisfying the nutritional requirements of plants [8]. The maintenance of this labile pool of available soil ^{137}Cs is also attributed to the cycling of organically bound ^{137}Cs and general lack of binding sites for fixation of Cs on mineral layer surfaces. These results support a hypothesis that Biochar added as a soil amendment may act to sequester labile soil ^{137}Cs in radioactivity contaminated coral soil from the Marshall Islands. Further studies will be required to determine the exact nature of the binding capacity of Cs on Biochar. The K_d experiments were conducted with soil water as the aqueous phase in an attempt to mimic field conditions for absorption of Cs onto Biochar in the environment. However, the behavioral and sorption properties of labile Cs in the environment may be affected by release of competing ions, and long-term mass-action, microbial, surface charge and/or other bio-geochemical reactions. Evidence of the true potential of Biochar as a remedial agent to reduce the soil-to-plant transfer of Cs isotopes will likely only come with field experimentation.

Conclusions

Initial tracer experiments designed to quantify the absorptive-desorptive capacity of Cs^{+} and Sr^{2+} ions on Biochar appear to demonstrate that this material does exhibit some favorable properties for fixation of Cs ions in suspended solutions of coral soil water. Measured ^{134}Cs $K_d$values on size fractionated Biochar after 7 days exceeded those exhibited by coral soil alone by factors of about 15–20 fold. The addition of Biochar as a soil amendment may therefore not only be beneficial in helping improve the fertility and water retention capacity of coral soil but provide a mechanism for sequestration of labile Cs ions that might otherwise transfer across root membranes to the fruiting body of plant foods. Conversely, there appears to be little or no added retention of Sr^{2+} relative to coral soil. Biochar may also act to help satisfy the nutritional requirements of plants and further reduce the soil-to-plant uptake of ^{137}Cs by providing a sustainable source of available K without the need for addition of large quantities of chemical fertilizers. Upon further research and testing, the use of Biochar for radionuclide remediation may not only be useful in the Marshall Islands but extend to other contaminated sites such as Fukushima.

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