Studying the radiogeochemistry of the Caribbean National Forest and the BONUS Power Plant in Puerto Rico

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The study of anthropogenic radionuclides has been conducted worldwide since the above and ground testing of nuclear weapon more than 6 decades ago. However, the island of Puerto Rico was used as living laboratory by government agencies and military purposes and there is no evidence of the current activities of the radionuclides release during the 1960's. This on going research has accumulated samples from the Caribbean National Forest (CNF) and the superheated prototype power plant BONUS in Rincon, PR. Gamma spectroscopy, grain size analysis and X-ray diffraction have been performed and the results showed the presence of Cs-137 in both sites.

Cs-137 activity at the CNF range from 0.361 Bq/g to below detection limits. Grain size analysis showed that the highest volume of clay at CNF was 27.17% and the lowest volume of clay was 6.84%. At the BONUS Reactor Cs-137 activity range from below detection limits to 0.011 Bq/g. In most cases clay volume was as high as 22.39% but also we found clay volumes as low as 2.79%. Major outcomes from this on-going investigation will include radionuclide concentrations, distributions and behaviors, with respect to aquatic geochemistry, dominant transport processes and ecological characteristics in Puerto Rico.

Introduction

- The Commonwealth of Puerto Rico, located in the Caribbean's Major Antilles, is comprised of several islands and its population is estimate to be over four million people.
- Puerto Rico's natural resources have been impacted by stress (e.g. viable drinking water limitations, increased erosion and sedimentation, decreased water quality, etc.), destruction (e.g. deforestation and habitat destruction); and anthropogenic contamination of the island's natural resources due in part by major development projects.
- Most rivers and reservoirs in Puerto Rico have been adversely impacted by man .
- Soil and groundwater near petrochemical facilities have been contaminated by accidental releases of petroleum, petroleum-related products and other chemicals, including benzene, toluene, ethyl benzene, and xylene .
- Heavy metals, trace elements and carcinogens have also negatively impacted Puerto Rico's natural resources.
- Puerto Rico's natural resources have also been exposed to anthropogenic radionuclides as a result of various US Department of Energy (US DOE) and Department of Defense (US DOD) sponsored activities.

Fig 1a. Map of Puerto Rico

Fig 1b. Map of BONUS Area in Rincon, PR

Boiling Nuclear Superheated Reactor

- During the early 1960's the US Atomic Energy Commission (US AEC) and the Puerto Rico Water Resource Authority (PRWRA) constructed the Boiling Nuclear Superheated (BONUS) reactor.
- The main purpose of this facility was to investigate the technical and economic feasibility of the integral boiling-superheated concept.
- The BONUS reactor experienced numerous problems, including 106 unintentional reactor shutdowns, one of which resulted in the released of 582.3 MBq into the atmosphere .
- BONUS was operational for a total of six years (1962-1968) before being decommissioned; a process which was completed in 1970.

Caribbean National Forest

- In the late 1960's the US DOE-sponsored the Puerto Rico Nuclear Center.
- The site was used to conduct experiments at the El Verde Experimental Station in the El Yunque Caribbean National Forest.
- Experiments involved inoculating and irradiating several trees with radionuclides.
- Radioactive Cs inoculation experiments were conducted on several trees in order to investigate the defoliation rates of various tree species.
- Results indicated that approximately 50 % of total amount of Cs-137 (17.02 MBq) injected into the trees was eliminated.
- It was also determined that the Cs-137 retained by the trees resulted in increased radiation levels of 200 mR/hr, in an area where the average background level was 5 mR/hr.
- During a set of experiments 777 MBq of tritium was injected into tree trunks in order to examine transpiration rates.

Study Area and Sample Collection

- Sediment cores from the island of Puerto Rico were obtained using a core liner.
- Sampling occurred at:
  - Caribbean National Forest, "El Verde" Experimental Station 18°19.351' N, 65°48.913' W
  - Bonus Reactor, Rincon 18°21.969' N, 67°16.095' W

•Cores were extruded, sectioned and transferred into bags, weighed, frozen and freeze-dried.

Gamma Analysis

- Samples were assayed for gamma emitters using a Canberra Genie multichannel analyzer planar gamma detector.
- Calibration was performed using US National Institute of Standards and Technology 4357 line and Ocean Sediment multi-line standards and the Canberra Multichannel MGS-5.
- Activities of the gamma emitters were determined via radionuclide-specific energy peaks located at 661 keV (Cs-137) and 46.5 (Pb-210).

Grain Size Analysis

- Analysis of sediment particle size was performed using the Saturn Digisizer 5200.
- To estimate the population size one gram of the freeze dried sample was sieved using a #230 mesh (62 µm) screen sieve.
- After sieving the sample, larger particles were retained.
- The grain size distribution of the smaller particles was determined using standard methods for the Saturn Digisizer and Saturn Software 5200.
- Particle size classes were defined as follows: clay size ≤ 4 µm; 4 µm < silt ≤ 63 µm; sand and larger > 63 µm.

Fig. 2a. Beach area at BONUS, Rincon, PR ; 2b BONUS Facility, Rincón, PR ; 2c Beach area ar BONUS, Rincon, PR. 2d. Inoculation Area, El Verde, Caribbean National Forest, Rio Grande, PR,; 2e Cocoa Beach, Rio Grande, PR; 2f Espiritu Santo River Area

X-Ray Diffraction

- Samples are treated for removal of organic matter, calcareous material and free Iron following Jackson 1958.
- Smear slides were prepared and analyzed using an XRD Commander for an hour period
- Each sample was reanalyzed after being treated with Glycol for 24 h in an effort to identfiy the expandable clays

Lost of Ignition Analysis

- Procedure was performed following Heiri et al. 2001 and Dean, 1974.
- Temperature was maintained at 105 o for 24 h, afterwards samples were ignited at 550 degrees for 4 h and LOI was calculated as follows:

$$LOI550 = ((DW\ 105 - DW\ 550) / DW\ 105) * 100$$

Results:

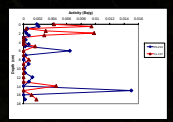


Fig. 3 Downcore Cs-137 activity at Coco Beach Estuary, PR. Errors associated with Cs-137 activities are ≤ ± 10 %.

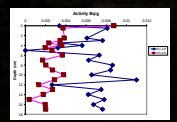


Fig. 4 Downcore Cs-137 activity at Bonus Area Station 1, Rincón, PR. Errors associated with Cs-137 activities are ≤ ± 10 %.

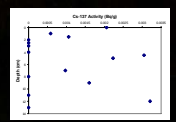


Fig. 5 Downcore Cs-137 activity at Bonus Area Station 2, Rincón, PR. Errors associated with Cs-137 activities are ≤ ± 10 %.

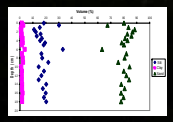


Fig. 6 Grain size distribution at Cocoa Beach, Rio Grande, PR.

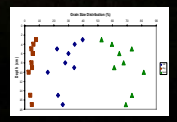


Fig. 7 Grain size distribution at Bonus Station 1, Rincón, PR

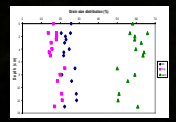


Fig. 8 Grain size distribution at Bonus Station 2, Rincón, PR

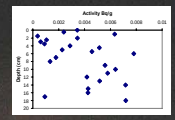


Fig. 9 Downcore Cs-137 activity at Bonus Area Station 7, Rincón, PR. Errors associated with Cs-137 activities are ≤ ± 10 %.

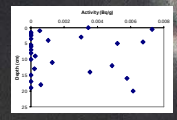


Fig. 10 Downcore Cs-137 activity at Bonus Area Station 6, Rincón, PR. Errors associated with Cs-137 activities are ≤ ± 10 %.

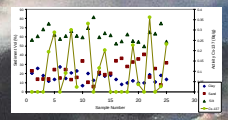


Figure. 11 Caribbean National Forest surface samples Cs-137 activity vs grain size distribution. Errors associated with Cs-137 activities are ≤ ± 10 %.

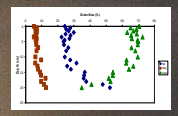


Fig. 12 Grain size distribution at Bonus Station 7, Rincón, PR

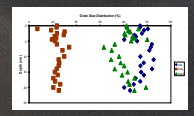
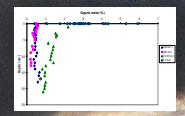
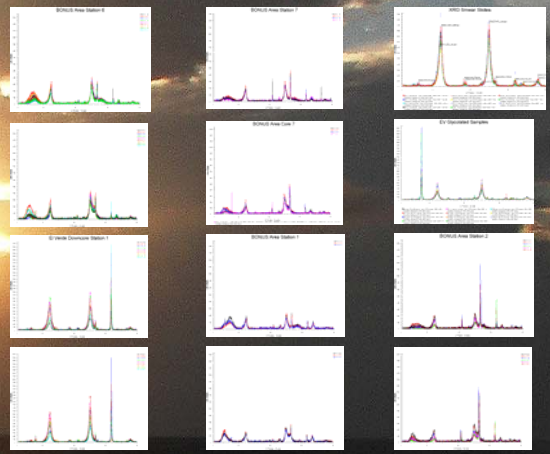


Fig. 13 Grain size distribution at Bonus Station 6, Rincón, PR





Preliminary conclusions:

- Sediment and soil samples indicate the presence of Cs-137 as well as Pb-210 at each study site
- The highest concentrations were determined for El Verde Surface samples. .
- Grain size analysis indicates that clay size particle average exceed 10 % at the Bonus Area, Rincon and is below 10% at El Verde and Espiritu Santo River .
- There is no strong correlation between particle size and Cs-137 retention at each site.
- XRD shows a peak that are homogeneous at all locations at El Verde Experimental Station surface samples as well as the BONUS Area.
- Kaolinite is present in all samples as well as chlorite.
- Organic content decreases with depth at sample sites and the highest content is in rainforest surface samples
- Further examination is needed to determine dominant factor(s) influencing the sediment retention capacity and stability at each location within Puerto Rico's ecosystem.

Acknowledgements:

We would like to acknowledge Dr. Joseph Smoak (Univ. of South Florida St. Petersburg), Ms.Nekesha Williams (Univ. of South Florida), Raquel Hernandez (Univ. of South Florida). Ms. Marietta Mayo (Univ. of South Florida), Kate T Ciembronowicz (USGS), Administration of Vieques National Wildlife Refuge and Oscar Diaz Refuge Manager, Dr. C. Benitez-Nelson (Univ. of South Carolina). This work was supported, in part, by the Univ. of South Florida Office of Sponsored Research New Researchers Grant Program Grant # R046908.

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