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Investigation of the dependence of the activity concentration of natural and artificial radionuclides on particle size in surface soil

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Abstract Surface soil radioactivity is being studied at NED-NTUA since almost thirty years. The last few years this research has expanded to the study of soil particle size fractionation of radionuclides. The purpose of the present work was to study the tendency of many radionuclides to accumulate in the smaller particle size fractions. To this end, soil samples were collected from an area in the NTUA Campus where ^{241}Am contamination, due to a failed ^{241}Am -tipped lightning rod, had been previously detected [1]. The samples were separated into size fractions using a sieving machine and analyzed by gamma spectrometry to determine ^{210}Pb , ^{241}Am , ^{234}Th , ^{228}Ra , ^{228}Th , ^{226}Ra , ^7Be , ^{137}Cs and ^{40}K . The activity concentrations of these nuclides were found to significantly differ among the size fractions examined.

Keywords soil, Americium 241, particle size, wet sieving

INTRODUCTION

The distribution of radionuclides in soil is influenced by many complex factors, among which are soil chemistry and radionuclide origin. Furthermore, it can be affected by several mechanisms depending on parameters such as the agent of water, ground inclination, wind intensity and even vegetation [2]. In previous research [3] it has been observed that variations in the activity concentration of radionuclides exist not only between different sampling points and sampling depths at the same location, but also between different particle size fractions within the same sample. In particular, many radionuclides were detected in higher concentrations in the smaller particle size fractions of soil, rather than the larger ones.

In the present work, further study is conducted to examine and obtain additional results on this particular tendency, including both natural and anthropogenic radionuclides.

SAMPLING AND SAMPLE PREPARATION

Surface soil samples were collected from an area where ^{241}Am contamination, due to a failed ^{241}Am -tipped lightning rod, had been previously detected [1]. Two samples with a mass of approximately 5 kg were collected from two different points, designated as points A and B, within a distance of 10 meters.

The samples were initially air-dried to <5% moisture. A standardized 282 cm³ container

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was filled with material from each sample for initial analysis, before any further processing. The samples were then dry sieved using a sieving machine equipped with sieves of standard sizes (45 μm , 63 μm , 125 μm , 250 μm , 500 μm , 1 mm, 2 mm). After sieving a standardized 97.2 cm^3 container was filled for each size fraction and sample. All containers were sealed with an epoxy resin to prevent the escape of radon and stored for approximately 1 month to attain radioactive equilibrium between ^{226}Ra and its short-lived decay products, before being analyzed using gamma spectroscopy.

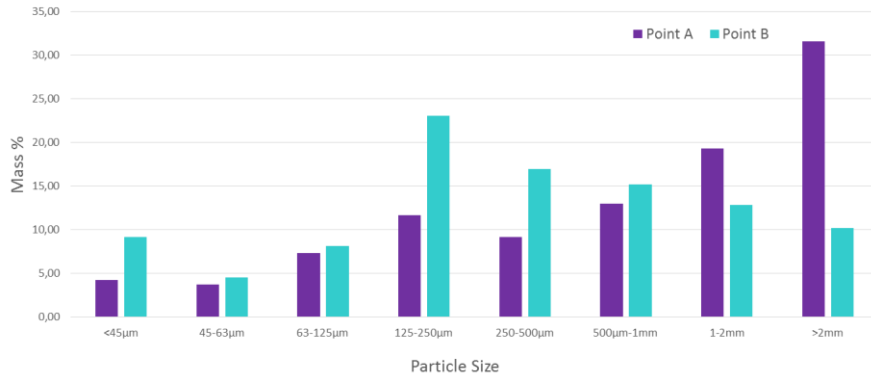


Fig. 1: Particle size analysis of soil samples determined using dry sieving

Visual observation of the samples indicated that the particles collected in the fractions > 1mm contain agglomerations of smaller particle sizes, rather than individual particles. In order to further investigate this finding, wet sieving analysis of the 1-2mm fraction of point A was performed, further subdividing the collected amount into four size fractions (<45 μm , 45-125 μm , 125-1000 μm , 1-2mm). For each one of the four size fractions, additional samples were prepared using 18.0 cm^3 containers. The samples were sealed and analyzed to compare with the corresponding size fractions obtained by dry sieving.

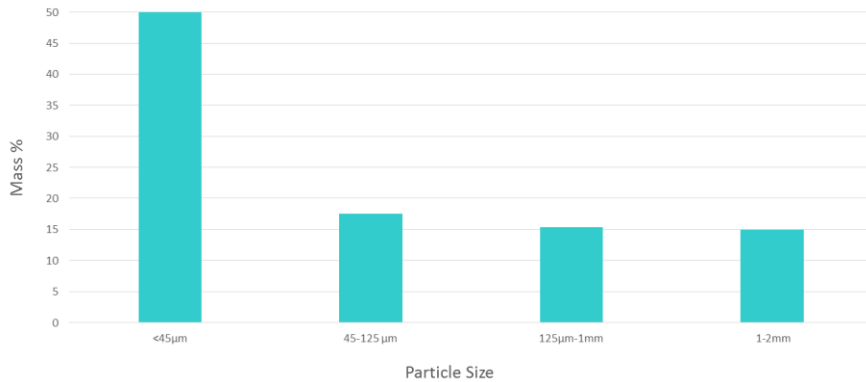


Fig. 2: Particle size analysis of the 1-2mm fraction of sample A determined using wet sieving

GAMMA-SPECTROSCOPIC ANALYSIS

The samples were analyzed using a planar LEGe detector and an HPGe detector. All spectra were analyzed using the SPUNAL code, developed in-house at NED-NTUA. The geometries of 282 cm^3 and 97.2 cm^3 have been calibrated experimentally, using a certified

mixed radionuclide solution, while the smaller geometry of 18.0 cm³ has been calibrated using the PENELOPE Monte-Carlo computer code [4]. Self-absorption corrections for the low energy photons were calculated via an experimental-numerical method developed at NED-NTUA [5].

RESULTS AND DISCUSSION

Analysis results for the fractions obtained by dry sieving are presented in Figures 3-7. Significant differences in activity concentration can be seen between the various size fractions of each sample in all cases. For most nuclides, activity concentrations tend to increase as particle size decreases.

It should be noted that significant differences can be observed for ²⁴¹Am and ²¹⁰Pb between corresponding fractions of the two samples, even though they have been collected within a distance of a few meters. This difference can possibly be attributed to water run-off from the adjacent building, as the sampling spots were selected in relation to drainage pipes.

Activity concentration results from the samples obtained by wet sieving are presented in Figure 8. It can be seen that significant differences exist, and in fact the results for the smallest size fraction are comparable to the values obtained from the corresponding dry sieving fraction. This indicates that particles in the 1-2mm size range are in fact agglomerations of smaller particles with inhomogeneous activity concentration.

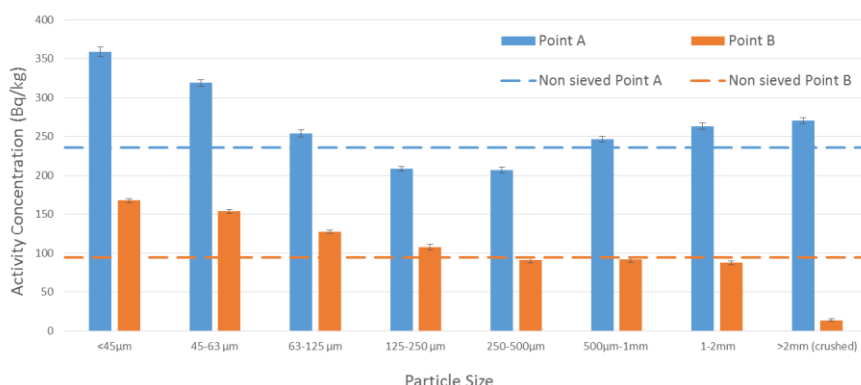


Fig.3: Activity concentration of ²¹⁰Pb in the dry sieving size fractions.

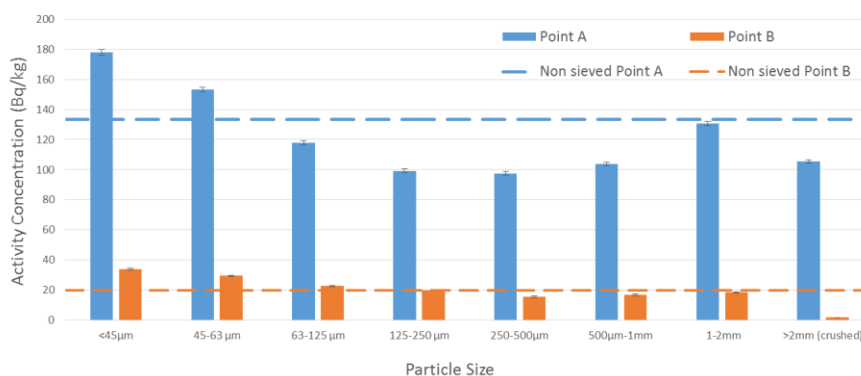


Fig.4: Activity concentration of ²⁴¹Am in the dry sieving size fractions.

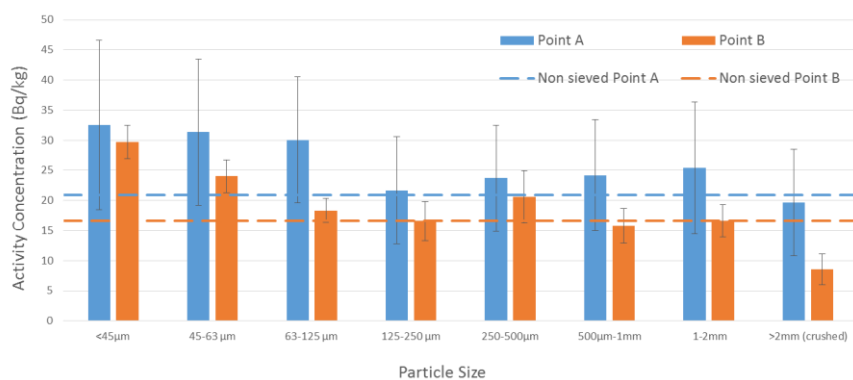


Fig. 5: Activity concentration of ^{234}Th in the dry sieving size fractions.

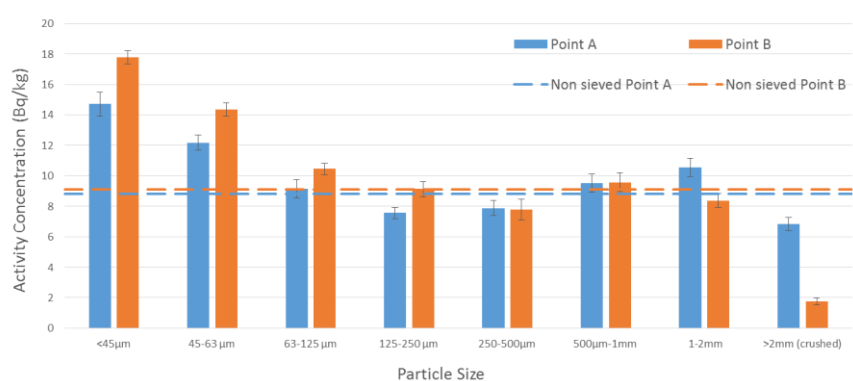


Fig. 6: Activity concentration of ^{137}Cs in the dry sieving size fractions.

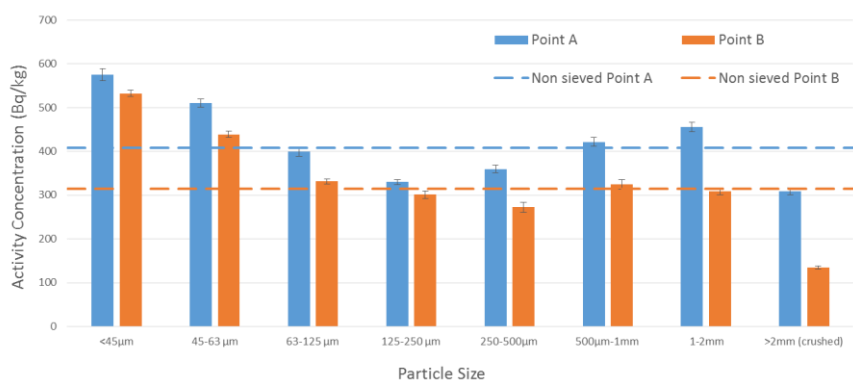


Fig. 7: Activity concentration of ^{40}K in the dry sieving size fractions.

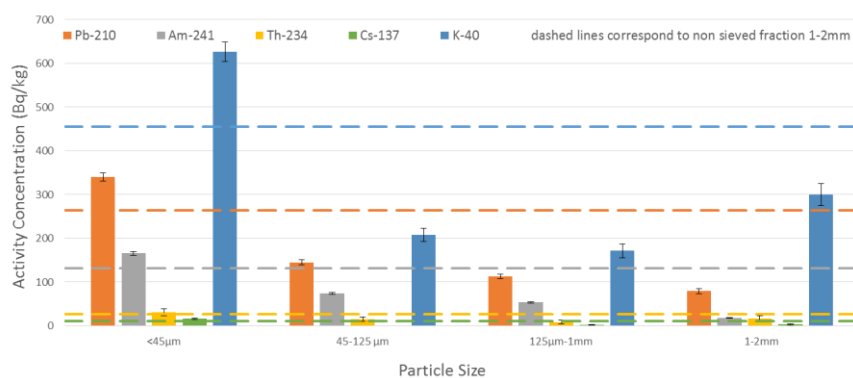


Fig. 8: Activity concentration of the size fractions of sample A obtained by wet sieving

CONCLUSIONS

Significant increase in the activity concentration of all radionuclides in the smaller particle size fractions of soil was observed, as expected from the literature [6]. For radionuclides deposited by fallout such as ^{137}Cs , ^7Be , ^{241}Am and ^{210}Pb this can be easily explained, since these radionuclides are deposited on the particle surface and the surface to mass ratio is higher for finer particles. However, for natural radionuclides originating within the soil matrix this trend is not as easily explained and should be further investigated. The fact that most radionuclides have increased concentration in the finer particles should be taken into consideration during sampling and sample preparation, as well as during radionuclides migration studies as well as soil erosion and sedimentation studies.

Significant variation in radionuclide concentrations was also observed even for samples collected within a few meters distance, while wet sieving results indicate that coarse particles may partly consist of agglomerated higher specific activity finer particles.

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