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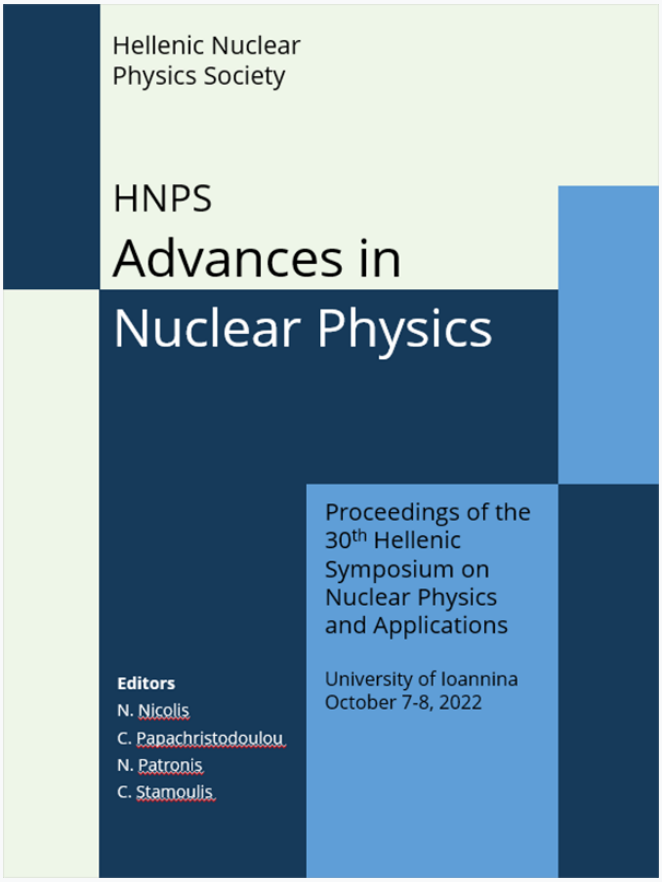

Surveillance of the seashore using the KATERINA II geo-referenced detection system

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Surveillance of the seashore using the KATERINA II geo-referenced detection system

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Abstract In this work, the design and initial demonstration of the KATERINA II detection system for rapid mapping of radionuclides in areas near to seashore is presented. A new development has been realized by integrating a GPS module in KATERINA II detection system and synchronizing its data with the acquired spectra in real-time. The new system may be used in a backpack, for areas with low activity concentration, or can be installed in an unmanned vehicle, for observing and mapping the source(s) of radioactivity, e.g. at the seashore, in areas with high contamination. A quantitative solution is provided for natural and artificial radionuclides, taking into account the characteristics of the detector, the parameters of measurement geometry and a mean beach sand/sediment composition. This paper reports field results for site characterization issues through automated analysis of gamma-ray spectra including low-level and low-energy γ-emitters. Perspectives of the future application of the system in a worldwide basis are related to radionuclides mapping and the assessment of dose rates in seashore areas that may be contaminated due to the operation of nuclear power plants, desalination plants and NORM industries, and/or due to the management of radioactive waste and the decommissioning of nuclear installations.

Keywords In-situ gamma-ray spectrometry; natural and anthropogenic radionuclides; total counting rate; beach sand; rapid mapping

INTRODUCTION

Ambient radioactivity is associated with terrestrial radionuclides as well as cosmic radiation and is the major source of human exposure to ionizing radiation that may result to doses which could reach a level as high as 80% of annual effective dose of an average person from the public. Humans may be exposed externally to γ-ray fields from natural radionuclides (^{238}U, ^{226}Ra, ^{232}Th and ^{40}K), as well as internally via inhalation of radioactive gases and airborne particles (^{222}Rn and its progenies) [1]. In radiological situations where long-term human/biota exposures and environmental impact are expected, the responsible authorities have to perform assessment of the affected areas. Furthermore, several incidences may take place in the vicinity of the shoreline due to anthropogenic activities (such as nuclear accidents, nuclear tests, and other processes that may release radionuclides into the environment). The above require the specific design of sophisticated environmental monitoring programs involving periodic and instant radiological surveys on a routine basis, as well as in cases of emergency. Fulfilling some of the objectives of the aforementioned programs, aiming to the radiation protection of the public and the environment, various environmental matrices such as sediments, coastal sands, and seawater have been studied during the last decades, to assess the state of marine environment in terms of radioactivity and related contamination and, in many cases, to produce radiological maps.

The mapping of beach sand is mainly performed either by a real-time and relatively fast *screening method* in the field, applying a radiometric scan with mobile mode of *basic* γ-spectrometric recordings, or by a lab-based *analytical method* which is comprised of the sampling strategy design and its implementation, the samples' preparation and their analysis by *advanced* nuclear spectrometric

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techniques. The main disadvantages of the lab-based method are that the radioactivity/radionuclidic maps are not produced rapidly; it is often laborious and not cost effective when time is limited and accuracy, precision and low detection limits are not the main objectives of a monitoring program, or decisions should be rapidly made in the field. Furthermore, the lab-based analytical method cannot be applied for precise hot spot localization since the number of samples is relatively limited; however, these spots can be identified with great precision by the mobile screening method. These drawbacks often limit the lab-based method to be applied when accuracy and precision are of great importance (e.g., for dosimetry assessments in crowded areas), or in emergency situations for radiation protection purposes (e.g., in case of an accident/incident), as well as to identify potential low level radioactive contamination from hot particles and other effluents in areas where nuclear installations operate or are under decommissioning.

In this work, the low resolution in-situ γ-ray spectrometry is optimized to perform surveys for rapid mapping of radioactivity using the geo-referenced KATERINA II system [2-5]. The scanning system was utilized to map natural radionuclides in addition to total counting rate of a specific area, as well as to identify potential areas with elevated radioactivity levels. Collected data analysis was performed using the R-code software platform. The tool (detection system and method) is tested in a region where radioactivity is at background level and a first quantitative mapping of natural radionuclides is presented.

EXPERIMENTAL DETAILS

The KATERINA II system is a γ-ray spectrometer widely used for marine applications [2-5]. It consists of a 3″x3″ NaI crystal, its power consumption is less than 1.0 W, and the spectroscopy as well as other operational parameters (such as high voltage, amplification gain, pole zero cancellation and baseline restoration) are adjustable by the embedded software. Energy resolution of the detector is 6.7% at the energy of 661 keV (^{137}Cs). The system can be operated in cabled mode (online) as well as in stand-alone and real-time mode, while it integrates externally a GPS getting continuously its position and logging in each spectrum the mid-position of the scanning track/path when moving (scanning mode). The scanning tracks follow the borders of an area of study, as well as the vertical and horizontal movements that should take place at low speed (1-2 km/h) within the area. Each spectrum contains the counts distributed in the respective energy channels and the lat-long coordinates (recommended preset time 20s). The maps are produced in a rapid manner using the R-language/code. In the near future, there will be an option for data transfer via mobile telephony network and WiFi. R-code is a language for statistical computing and graphics and may implement a wide variety of statistical (linear and nonlinear modelling, classical statistical tests, time-series analysis, classification, clustering and others) as well as graphical methods and techniques. R-code is available as free software that can be compiled and run on a wide variety of computer platforms (UNIX/Linux platforms and similar systems, Windows and MacOS).

The system was demonstrated in the frame of a scanning/mapping benchmarking, under the auspices of IAEA, in the Attica region close to a past mining area named Limani Passa, Lavrio, Greece (Fig. 1). This is a preliminary study to depict in quantitative terms the activity concentration of key natural radionuclides for performing radiological studies, as well as for studying the coastal variability in areas that are affected from anthropogenic activities and natural processes (such as climate change). The first step of analysis was to provide the spatial distribution of the total counting rate by summing up all the recorder gamma-ray counts (along transects) and dividing by the spectral data acquisition duration.

Figure 1. *The study area (the beach is named Limani Passa, Lavrio and the area is depicted with the pin)*

SPECTRA ANALYSIS & SIMULATION SET UP

After scanning completion, an automated analysis of each 20s-acquired spectrum was performed using the energy windows of ^{40}K (1360 - 1560 keV) at the centroid of 1461 KeV, ^{214}Bi (1650 - 1870 keV) at the centroid of 1764 keV (^{238}U series) and of ^{208}Tl (2480 - 2740 keV) at the centroid of 2614 keV (^{232}Th series). The massic activity (Bq/kg) of each one of the above radionuclides was roughly determined via the count rate in its respective window divided by calibration factors that were calculated using EGSnrc MC Simulation code [6].

The modeling of KATERINA II system was implemented on the base of the real-detailed geometry and chemical composition of its components. The γ-photons emitting sandy beach surface was modeled as a cylinder of 6 m diameter and 0.3 m in height, consisting of a 1.7 kg/L homogeneous matrix containing 8.38% C, 54.54% O, 6.77% Al, 10.17% Si, 0.44% S, 2.23% Cl, 0.63% K, 9.41% Ca, 7.03% Fe, 0.19% Sr and 0.21% Pb (empirical composition). The distance between KATERINA II frontal window (downlooking) and beach surface (upper cylinder base) was set at 1 m, as it is the case during scanning (minimum 2 m away from the sea-front). Based on the fact that the simulated photon energies are relatively high (661, 1461, 1764 and 2614 keV), the body of the operator was not modeled since no significant attenuation was expected according to the preliminary study. The simulation set up is depicted in Fig. 2.

For each one of the energies of interest, 10 batches x 5.000.000 “histories” of photons generated in the sand-source were simulated and analyzed. NIST cross-section data libraries were used for the simulated photon/electron-matter [7]. The calculated efficiencies are the calibration factors that were used to provide quantitative data for the maps. In addition to the efficiencies, the influence of the scattered 2614 keV photons to the continuum at ^{214}Bi and ^{40}K windows was calculated for subtraction purposes. The same was applied regarding the influence of the scattered 1764 keV photons to the continuum at ^{40}K window.

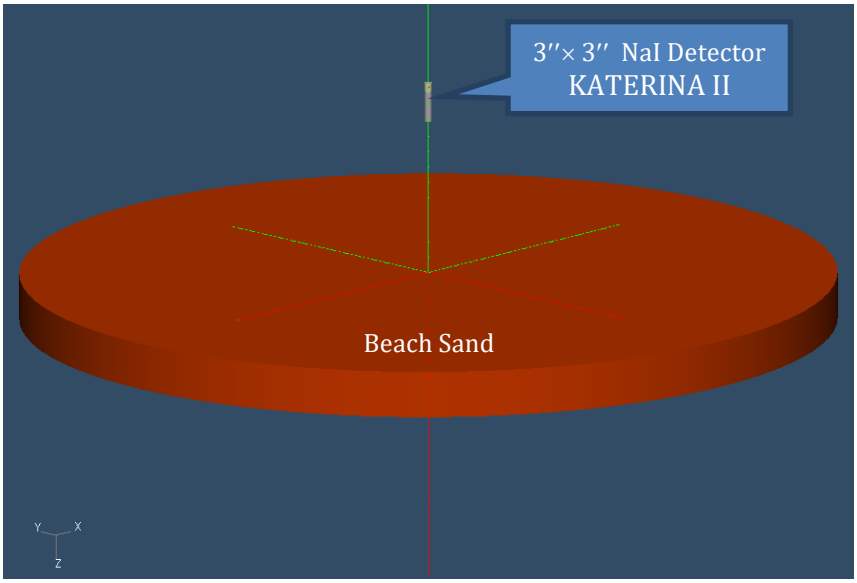

Figure 2. *Illustration of the simulation set up for the measurement geometry.*

RESULTS AND DISCUSSION

Maps of radioactivity

The spatial distribution of the activity concentration (Bq/kg) of ^{40}K on Limani Passa beach is depicted in Fig. 3. The activity concentration varies from 50 to 300 Bq/kg (1.6 - 9.5 g elemental K/kg of sand) along the shoreline. The variation of ^{40}K along the beach is due to the fact that the granulometry of the sand exhibits differences since the fine grains have higher concentration of ^{40}K. The two edges of the beach are affected from two streams and this may probably change the concentration of ^{40}K due to erosion processes. In Fig. 4 the spatial distribution of ^{208}Tl (daughter of ^{232}Th series) is also depicted. The counting rate at the energy window of 2614 keV (emission energy of ^{208}Tl) is generally very low and all recorded counts at this region of interest are considered for the analysis procedure. The results of the activity concentration of ^{208}Tl varies from 2 up to 18 Bq/kg due to the fact that the granulometry of the sand is not homogeneous and that the edges of the beach are affected from streams that create a seasonal coastal variability of the shoreline caused by the suspended matter that is deposited to the specific area. Finally, ^{214}Bi distribution is illustrated in the map of Fig. 5. For mapping purposes, the assumption of ^{238}U/^{226}Ra/^{214}Bi equilibrium is considered [8, 12]. Additionally, laboratory analysis of 10 sand samples (two measurements/sample, with the 1st one taking place immediately after airtight sealing and the 2nd one ~30 days later) verified that the exhalation of ^{222}Rn from Limani Passa sand was negligible (approx. at a level in the range of 5-15%). According to the above assumption, the ^{238}U massic activity in Limani Passa beach sand varies within the range 1 - 38 Bq/kg (0.081 - 3.1 mg elemental U/kg of sand). This massic activity range is in agreement with ^{238}U concentration in earth crust materials, as determined by numerous other research groups around the world [8-14]. Also, the massic activities of ^{238}U progenies estimated in this work are within the range of ^{238}U concentration in soils found in Greece (1 - 240 Bq/kg) [10, 12]. In terms of average values, the total of acquired scanning data regarding ^{238}U daughters, enable the low-resolution in-situ gamma-ray spectrometry to quantify activity concentrations even as low as the background level (this is also the case in the area of study). Furthermore, the uncertainties of the measurements can be enhanced compared to the lab-based method. For instance, in case of the analysis of twenty-five spectra acquired in neighboring points of the beach (total acquisition time 500s), the relative uncertainty of ^{208}Tl, ^{214}Bi and ^{40}K is 8, 20 and 18%, respectively, as resulted from the statistical analysis of the photopeak areas. Finally, the uncertainty

budget should be determined and evaluated applying the method in different coastal systems. In case of highly contaminated areas, the uncertainty values will be drastically reduced due to the improved counting statistics.

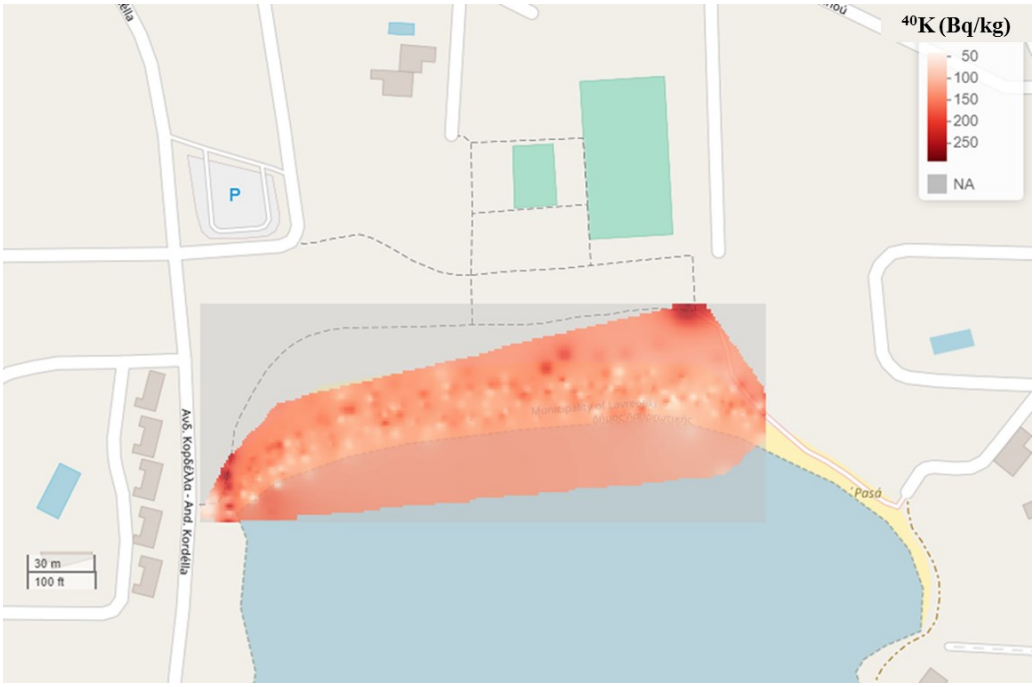

Figure 3. *The spatial distribution of the activity concentration (Bq/kg) of ^{40}K in Limani Passa beach*

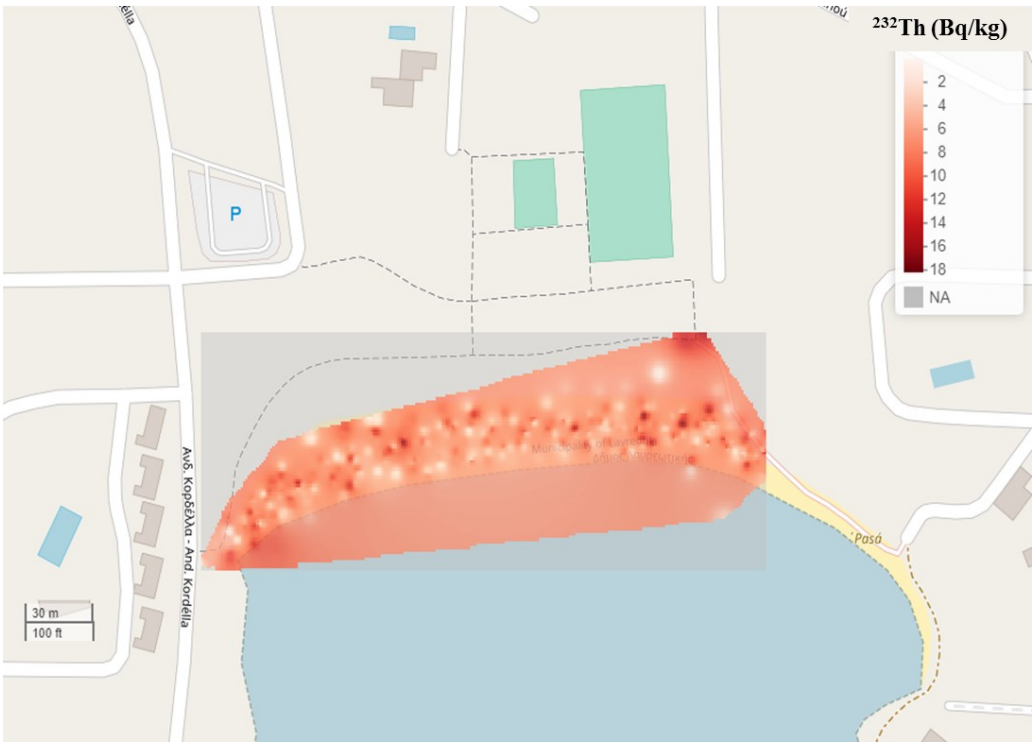

Figure 4. The *spatial distribution of the activity concentration (Bq/kg) of ^{208}Tl (^{232}Th series) in Limani Passa beach*

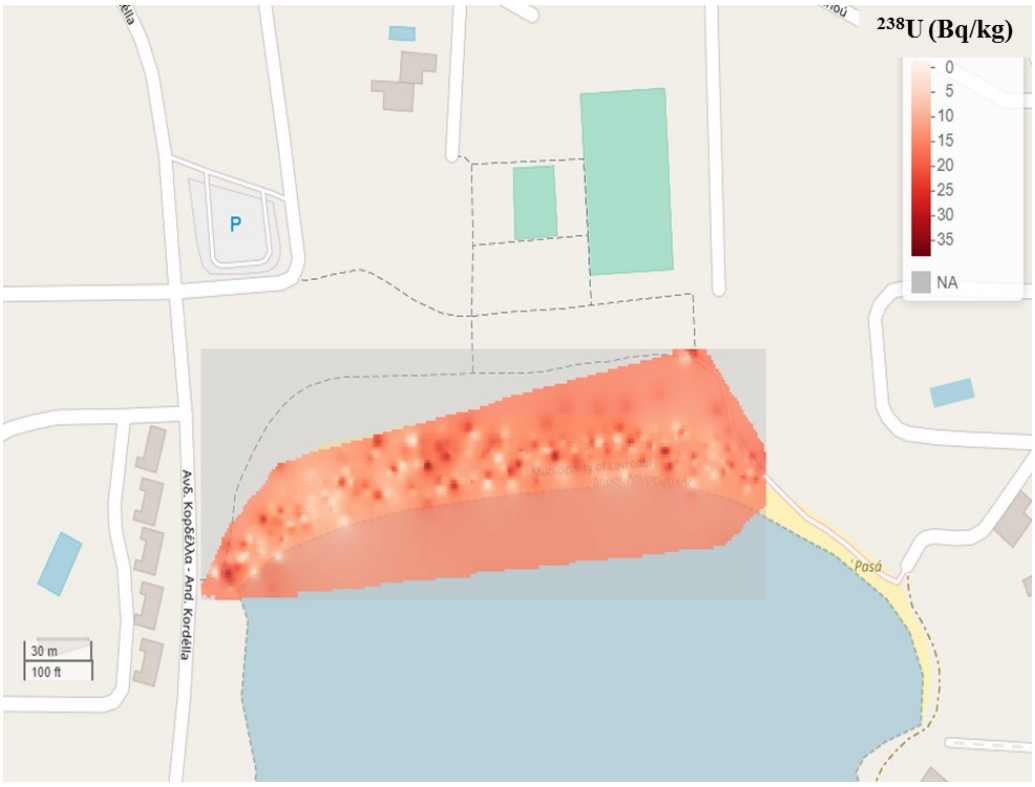

Figure 5. *The spatial distribution of the activity concentration (Bq/kg) of ^{214}Bi (^{238}U series) in Limani Passa beach*

Validation/Inter-comparison

Aiming to validate the described quantification method, by examining the correlation between the MC-calibrated mapping results and the exact activity concentration of NORs (Natural Occurring Radionuclides) at specific locations on the scanned beaches, 10 sand samples were collected. The samples were dried at 105 ºC (overnight) and sieved after homogenization. A portion from each sample was transferred into an in-lab designed and custom produced cylindrical container of optimized geometry and material, pressed up to the standard counting geometry (105 ml) with the aid of a special pressing tool, immobilized by a circular polymer-coated aluminum sheet and weighed. The containers were sealed with the use of epoxy resin for ^{222}Rn confinement. Samples' final analysis was initiated when ^{226}Ra/^{222}Rn/^{214}Pb/^{214}Bi and ^{228}Th/^{224}Ra/^{212}Pb/^{208}Tl series reached equilibrium (~30 days after sealing). Low-level γ-ray spectrometric analyses of the sand samples were performed using a broad energy HPGe detector with Be window, which is 4π-shielded by a 11cm Lead (Sn-Cu lined) shield (product of teleDOS Nuclear Tech). Due to the relatively low NORs' activity concentrations in the specific samples, the duration of spectra acquisition was set at 300000 s for better statistics. Spectra analysis was performed, after spectrometer-samples system calibration, using the Genie 2k suite software.

The calibration of the system was implemented via the standard lab method of teleDOS Laboratories; i.e. using the EGSnrc MC code taking into account the density of each sample, an average sand composition as above (for KATERINA II-beach modelling), as well as the exact geometry of the measurement and the detector. The calibration was evaluated, and Quality Control was applied using Certified Reference Materials of similar composition and physical properties with the samples. Moreover, the Analytical Quality of the lab is periodically evaluated (www.teledos.eu/PT).

The MC-calibrated mapping results in the specific 10 locations of the beaches were found to be in good agreement with the lab-based measurements of the activity concentration of NORs in the respective samples as determined through high resolution γ-spectrometry: the minimum and maximum

bias between the values of the massic activities as determined by the two methods were found between 2% to 18% for each NOR of interest. However, an optimization of the quantification method and uncertainty budget analysis is required for better accuracy and higher precision of the in-situ method.

CONCLUSIONS

The proposed method provides in a quantitative manner the spatial distribution of the activity concentration of natural radionuclides (NORs) in matrices like beach sand. The γ-scanned area can be as large as needed since the survey is relatively fast, without compromising spatial resolution, the geo-referenced KATERINA II system is weather-resistant, and its autonomy is long enough to map several areas/places at once. The maps are produced rapidly, immediately after the experiment, and further analysis can be performed (guided by the initial spatial variation) taking e.g. into account the maximum and minimum values of the activity concentration of a nuclide of interest. The proposed geo-referenced method was validated using the lab-based high-resolution gamma spectrometry method, taking into account the measurement geometry and the elemental composition of the beach sand samples. The in-situ screening method for mapping terrains like beaches is optimized for quantitative results regarding NORs (^{40}K, ^{238}U series and ^{232}Th series) using the corresponding energy windows where the key γ-photons emitted by the daughters of the above series and potassium are detected. In the future, the quantification process will be improved performing long term scans of longer durations in order to improve statistics and including practices of uncertainty budget analysis.

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References

[1] UNSCEAR 2010. Sources and Effects of Ionizing Radiation - Exposures of The Public and Workers from Various Sources of Radiation - UNSCEAR 2008 Report, United Nations Publication (2010).
[2] C. Tsabaris et al. Ninth International Symposium on Monitoring of Mediterranean Coastal Areas: Problems and Measurement Techniques, Livorno, Italy (2022)
[3] C. Tsabaris et al., International Conference on the Safety and Security of Radioactive Sources: Accomplishments and Future Endeavors (CN-295), IAEA, Vienna, Austria (2022)
[4] C. Tsabaris et al., Second International Conference on Applications of Radiation Science and Technology (ICARST-2022), IAEA, Vienna, Austria (2022)
[5] Alexakis et al., J. Mar. Sci. Eng. 9, 910 (2021)
[6] The EGSnrc Code System: Monte Carlo Simulation of Electron and Photon Transport. I. Kawrakow, E. Mainegra-Hing, D.W.O. Rogers, F. Tessier, B.R.B. Walters. NRCC (2015)
[7] XCOM: Photon Cross Sections Database. M.J. Berger, J.H. Hubbell, S.M. Seltzer, et al., NIST, PML, Radiation Physics Division (URL: https://dx.doi.org/10.18434/T48G6X).
[8] IAEA TECDOC-1363. Guidelines for radioelement mapping using gamma ray spectrometry data (2003)
[9] UNSCEAR 2008 Report - Sources and Effects of Ionizing Radiation. Volume I. United Nations Publication (2010)
[10] UNSCEAR 2000 Report - Sources and Effects of Ionizing Radiation. Volume I: Sources. United Nations Publication (2000)
[11] I. Pais, J.B. Jones. The Handbook of Trace Elements (1997)
[12] M.J. Anagnostakis, E.P. Hinis, S.E. Simopoulos. 7th International Symposium on The Natural Radiation Environment (NRE-VII), Radioactivity in the Environment Vol. 7 (2005)
[13] K. Froehlich. Environmental Radionuclides: Tracers and Timers of Terrestrial Processes, Radioactivity in the Environment Vol. 16 (2010)

[14] A.S. Paschoa, F. Steinhauesler. Technologically Enhanced Natural Radiation, Radioactivity in the Environment Vol. 17 (2010)