



DOSE RATE ASSESSMENT OF ^{137}Cs TO PELAGIC FISH USING AN INNOVATIVE METHOD COMBINING FIELD MEASUREMENTS, CMEMS DATA AND ERICA ASSESSMENT TOOL

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Abstract. Earth Observation satellites and environmental models are able to monitor changes of ecological parameters in the marine environment. Radionuclides cannot be directly measured using satellite remote sensing, because they are not currently detectable by the satellite instruments. Nevertheless, the levels of radionuclides, in the marine environment are known to be associated with physical and biogeochemical parameters of the natural environment. Considering this attribute, we investigate the potential relation between ^{137}Cs activity concentration and sea surface salinity. We select the parameter of salinity, as the element of Cesium in the seawater is conservative and contributes to it. Cs-137 activity concentration measurements are issued from the database of the Environmental Radioactivity Laboratory (ERL) of NCSR “D” for the period 1993 to 2006. Salinity corresponds to sea surface salinity (SSS) data issued from Copernicus Marine Environment Monitoring Service (CMEMS) database spanning also the period 1993 to 2006. A total of 15 measurements are used for the establishment of a linear regression model for the marine environment of the Island of Lemnos (Greece). The Island of Lemnos is located in the Aegean, southwest of the Dardanelles Strait and its waters are constantly enriched with ^{137}Cs of Black Sea origin. The resulting linear model ($R^2=0.82$) is then validated using recent ^{137}Cs measurements spanning November 2018 and July 2019. During two sampling cruises that took place, on 12-15 November 2018 and on 24-28 July 2019, a total of 11 samples were collected and analyzed. The measured concentrations obtained by gamma spectrometry, in terms of activity concentrations (Bq/m^3), are then compared with the estimated ^{137}Cs concentrations obtained by the model. The estimations present a relative error of less than 25%. Finally, in order to conduct the risk assessment in the studied area, the dose rate thematic maps in the marine area of Lemnos are calculated with the ERICA Assessment Tool and QGIS for pelagic fish, as one of the most representative organism of the studied environment and the most important, in terms of commercial value. The doses in pelagic fish are calculated for each pixel within the estimated ^{137}Cs activity concentrations thematic maps for November 2018 and July 2019. The results show the corresponding dose rate maps for the pelagic fish during November 2018 and July 2019 for the Lemnos Island. The dose rates in the thematic maps vary from 0.7 to 1.1 $\mu\text{Gy}/\text{year}$, which are far lower than the intervention levels, indicating low impact due to the ^{137}Cs exposure.

Keywords: Dose rates, pelagic fish, marine radioactive pollution, ^{137}Cs activity concentrations, CMEMS, ERICA Assessment Tool, Aegean Sea, Lemnos Island

1. INTRODUCTION

An innovative system is created for radiological risk assessment in the area of Lemnos Island. Lemnos Island is located in the Eastern Mediterranean, South Aegean Sea and its waters are constantly enriched with ^{137}Cs of Black Sea origin [1, 2, 3]. In the Eastern Mediterranean area, ^{137}Cs mainly originated from atmospheric fallout (nuclear weapon tests, 1945 - 1960) and the Chernobyl reactor accident in 1986 [2, 3].

The fate of radionuclides in the marine environment, as the marine environment is a dynamic system, is not to stay in a steady state condition and area but to be transported and dispersed both horizontally and vertically [4]. According to their physicochemical properties and their behavior in the seawater, radionuclides are categorized to conservative and non-conservative. Conservative radionuclides like ^{137}Cs , have a low capacity for adsorption onto particles, have long residence times, their mass is conserved in water making their

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scavenging from the aqueous phase very slowly and they are mainly present in ionic form (Low molecular mass - LMM) making them more bioavailable to organisms [2, 5, 6, 7]. Due to their solubility, conservative radionuclides are not able to be monitored using Earth Observation (EO) satellites and environmental models. However, radionuclides in the marine environment are known to be associated with physical and biogeochemical parameters of the natural environment. Considering this attribute, we investigate the potential relation between ^{137}Cs activity concentration and sea surface salinity (SSS), as the element of Cs in the seawater is conservative and contributes to it. The relation of ^{137}Cs activity concentration and SSS, will result in a model crucial for the creation of a multidisciplinary risk assessment system for Lemnos Island.

Radiological risk assessment for pelagic fish is important in the Lemnos marine area, as its waters are currently and constantly enriched with ^{137}Cs and due to the oceanographic conditions is one of the most productive areas in the Aegean Sea. Lemnos Island has a lot of fisheries, fishery facilities and professional fishermen with boats [8, 9, 10].

The objectives of this study are the following: (i) to examine the relation of ^{137}Cs activity concentration from the database of the Environmental Radioactivity Laboratory (ERL) of NCSR "D" and SSS from Copernicus Marine Environment Monitoring Service (CMEMS) for the creation of a linear model using ^{137}Cs activity concentration and SSS data from 1996-2006, (ii) to validate the resulting linear model with new ^{137}Cs measurements spanning November 2018 - July 2019, and (iii) to conduct the risk assessment in the marine area of Lemnos island using the ERICA Assessment Tool and QGIS for pelagic fish.

2. STUDY AREA

Lemnos Island is located in the North Aegean Sea (NAS) in the Eastern Mediterranean (Figure 1). NAS has the widest continental shelf, is the most productive region of the Aegean Sea and the most important area for the Hellenic fisheries. NAS is characterized by relatively high levels of productivity, abundant fish stocks (especially small pelagic species) but also high biological and hydrological heterogeneity [8, 9, 10].

The circulation in the area is mainly determined by the surface inflow from the Dardanelles, which, under the influence of the prevailing wind patterns, shifts above or below the island of Lemnos, creating various thermohaline fronts and gyres [11, 12]. The climate in the area is characterized by a typical Mediterranean type of climate with Sea Surface Temperature values from a minimum of 15 °C in February to a maximum of 26 °C in August [13]. SSS values vary from 31 to 39 psu, with bigger salinity fluctuations present during the summer, while during the winter the salinity is mostly stable [14].

The major sources of ^{137}Cs to NAS are the global fallout from nuclear weapon testing, mainly in the period 1945–1960 and the Chernobyl accident in 1986 [2, 3]. In 1986 the average deposition of ^{137}Cs from the fallout was estimated to be approximately 4 kBq m⁻². The total radiocesium input from Chernobyl fallout in the Black Sea and the Eastern Mediterranean has been

estimated to be 2400 TBq for the Black Sea and 820 TBq for the Aegean Sea. After 2000, the radiological status of the NAS is characterized by mean ^{137}Cs activity concentration 5 ± 0.5 to 13.3 ± 1.3 Bq m⁻³ [2, 3].

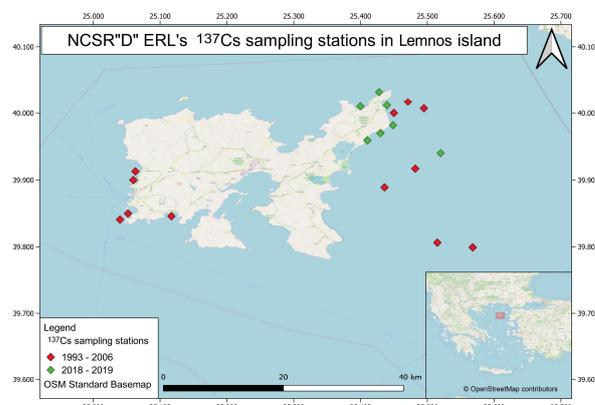


Figure 1. NCSR"D"/INRASTES/ERL's ^{137}Cs sampling stations in Lemnos Island. Red: Stations active from 1993-2006. Green: Stations active from 2018-2019. Basemap from OpenStreetMap (OSM) contributors [15]

3. METHODOLOGY

The methodology for the completion of this study includes sea water samplings and laboratory analyses for ^{137}Cs activity concentration, SSS data retrieval from CMEMS, linear regression analysis, ^{137}Cs activity concentration estimation and validation and dose rate assessment in pelagic fish (Figure 2).

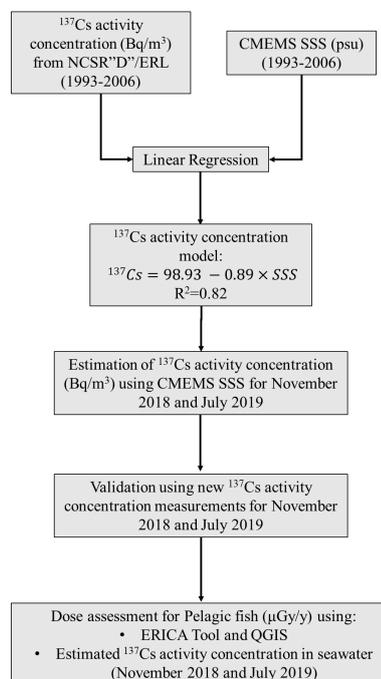


Figure 2. Flowchart of the methodology

3.1. ^{137}Cs activity concentration data

The ^{137}Cs activity concentration data concerning the regression analyses were obtained for the NCSR"D"/INRASTES/ERL database for the years

1993-2006 (Figure 3). These data (15 measurements) derived from the ERL's sampling stations in NAS where sampled seawater (volume of 60L or 1000L according to laboratory analysis method) was analyzed either using the Ammonium Phosphomolybdate (AMP) method or the Copper Ferrocyanide filters method and then measured with gamma spectrometry [2, 3].

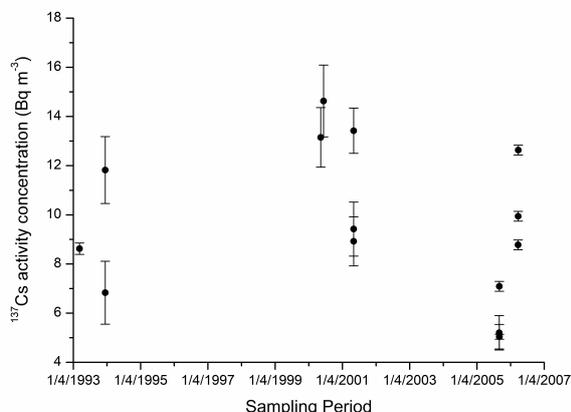


Figure 3. ^{137}Cs activity concentration for 1993-2006 [2]

The ^{137}Cs activity concentration data for the validation were obtained by 2 sampling cruises in Lemnos Island during November 2018 and July 2019. Where using a table top sampler and filters impregnated with Copper Ferrocyanide, a total of 11 samples were collected and transferred to the ERL for laboratory treatment and gamma spectrometry measurements.

Both the AMP and the copper ferrocyanide methods are based on the ion exchange of ^{137}Cs with chemical compounds and resins [2, 3, 16, 17]. In the first method 60L of seawater are analyzed using the yellow reagent AMP $[(\text{NH}_4)_3\text{P}(\text{Mo}_3\text{O}_{10})_4]$ along with ^{134}Cs as a carrier and yield tracer. While in the second method using water filters impregnated with copper ferrocyanide $(\text{Cu}_2[\text{Fe}(\text{CN})_6])$, 1000L of seawater is passed through for in situ adsorption. The results of each one of these methods are then placed in a calibrated measurement pot (radius 3.4cm, height 2cm) and measured with gamma spectrometry.

The gamma spectrometry measurements are carried out using a Canberra system comprising of a High Purity Germanium (HPGe) detector system with an efficiency of 90% (relative to a $3''\times 3''$ NaI(Tl) crystal) and resolution of 2.1 keV at the 1.33 MeV photopeak of ^{60}Co ; that is energy calibrated using a multi gamma standard source covering an energy up to 2000 keV. The efficiency of the detector is calculated using the same standard active source under the same geometry with the pot used for the measurement of the samples [2, 3]. The software used with the detector is Genie 2000. The samples are then measured for 72 hours. The activity concentration of the sample is calculated in Bq/m^3 of seawater [2, 3]. The gamma spectrometry measurements in ERL are certified each year by participating in the ALMERA network (Analytical Laboratories for the Measurements of Environmental Radioactivity) intercalibration from IAEA and also they are accredited by the Hellenic Accreditation System (certificate no. 447-4 to ELOT EN ISO/IEC 17025:2017).

3.2. CMEMS SSS data

The SSS parameter from the CMEMS marine database derives from marine environmental models and is validated using in situ buoy data [18]. In this study the data products-datasets MEDSEA_MULTIYEAR_PHY_006_004 and MEDSEA_ANALYSISFORECAST_PHY_006_013 are used. The model thematic maps cover the entire Mediterranean Sea, with a spatial resolution of about 4km (0.042deg x 0.042deg). For the regression purposes, 15 SSS measurements spanning the years 1993-2006 (corresponding to the same dates as ^{137}Cs measurements) are used from the MEDSEA_MULTIYEAR_PHY_006_004. While for the validation purposes, 11 measurements for November 2018 and July 2019 (corresponding to the same dates as ^{137}Cs measurements) are used from MEDSEA_ANALYSISFORECAST_PHY_006_013. Moreover, for the QGIS platform 2 monthly SSS maps for November 2018 and July 2019 are used from MEDSEA_ANALYSISFORECAST_PHY_006_013.

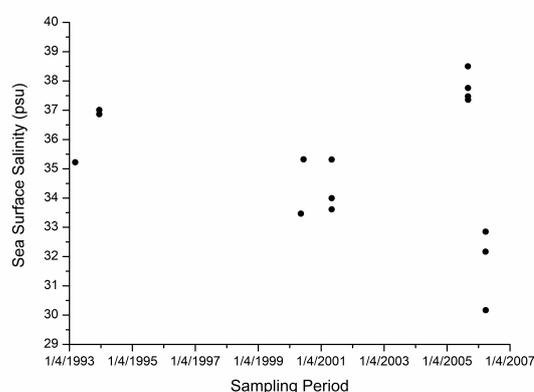


Figure 4. SSS from CMEMS for 1993-2006 [18]

3.3. Regression analysis

Statistical regression analysis is performed by single linear regression using SSS as the independent variable and the ^{137}Cs activity concentration as the dependent variable. The regression is performed for the years 1993-2006 using 15 conjoined measurements of the independent and the depended variables. From this regression a linear model for the estimation of ^{137}Cs activity concentration from SSS is derived.

3.4. Model Validation

The validation of the linear model is performed by estimating the ^{137}Cs activity concentration using CMEMS SSS data (November 2018-July 2019). Afterwards, the estimated ^{137}Cs data are compared to the measured ^{137}Cs data (11 measurements) for November 2018 and July 2019. To this purpose, we first calculate the absolute estimation error for each pair of measured and estimated ^{137}Cs values, as seen in equation (1).

$$\text{Absolute estimation error} = \frac{|^{137}\text{Cs}_{\text{est}} - ^{137}\text{Cs}_{\text{meas}}|}{^{137}\text{Cs}_{\text{meas}}} \quad (1)$$

Where, $^{137}\text{Cs}_{\text{est}}$ is the estimated ^{137}Cs activity concentration and $^{137}\text{Cs}_{\text{meas}}$ is the measured ^{137}Cs activity concentration.

Then, the relative absolute estimation error is calculated, which corresponds to the average of all the absolute estimation errors.

3.5. Risk Assessment

The risk assessment in pelagic fish, is performed using the ERICA Assessment Tool and the QGIS software.

The ERICA Assessment Tool (version. 1.3.1.51, 12/07/2019) is applied based on the estimations of radionuclide activity concentration that were calculated for November 2018 and July 2019 [19]. The total absorbed dose rate that incorporates the external and internal exposure to radionuclides is calculated by the modeled activity concentration of ^{137}Cs in sea water. Taking into consideration the ecological characteristics of the specific ecosystem, the risk analysis in this region is performed based on the most abundant species found in this region, the pelagic fish, as the generic phantom organism of the tool that represents the common sea-water fish is called.

QGIS is a freeware GIS where it can be used for mapping and geospatial applications [20]. After the calculation of the dose rates for pelagic fish, the data are then inserted to the QGIS (version 3.18.3 with GRASS 7.8.5, 15/05/2021), and mapped using the longitude and latitude values in the 1984 World geodetic system (WGS 84 or ESPG:4326). Then the vector points are transformed to a grid with an extent matching the estimated ^{137}Cs activity concentrations using the Geographically Weighted Regression (GWR) tool in the SAGA toolbox, which is the most successful spatial interpolation method for environmental studies as it calculates the dispersion of a parameter using another closely linked one (in this case dose rate and ^{137}Cs activity concentrations) [20, 21, 22]. The final dose rate maps for pelagic fish are for November 2018 and July 2019.

4. RESULTS AND DISCUSSION

The results of this study include the regression results and the model for the ^{137}Cs activity concentration estimation, the validation of the model using new measurements in the Lemnos Island and the dose rate calculations for pelagic fish.

4.1. Regression-Linear model

The relation of ^{137}Cs activity concentration and CMEMS SSS seems to be best described by an inverted linear model. The resulting model has an R^2 value of 0.82 indicating a strong relation between the ^{137}Cs activity concentrations and the SSS. The resulting equation (2) is:

$$^{137}\text{Cs} = 98.93 - 0.89 \times \text{SSS} \quad (2)$$

The negative correlation in this specific area of NAS is due to seawater characteristics and the seawater circulation. The Black Sea waters that are richer in ^{137}Cs and less saline are constantly mixed with the Aegean Sea waters that are poorer in ^{137}Cs and more saline [1, 2, 23].

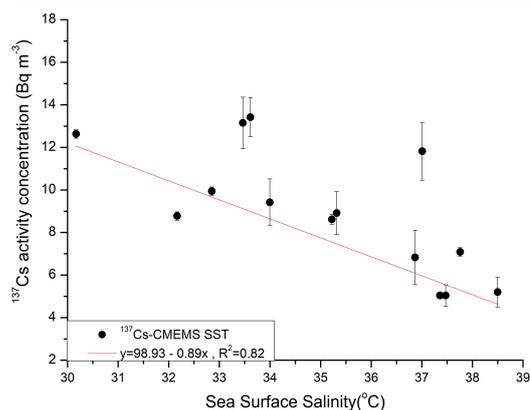


Figure 5. Regression of ^{137}Cs activity concentration and CMEMS SSS in Lemnos Island during 1993-2006.

4.2. Validation

Concerning the model validation process, the modeled-estimated ^{137}Cs values are close to the measured ^{137}Cs values presenting a relative absolute estimation error of 35%. Apart from the relative absolute estimation error, the modeled data also seem to be in range with the measured data, presenting ^{137}Cs activity concentration values from 4.5 to 8.0 Bq/m³.

Moreover, the model seems to correspond well in the different seasonal periods, where during the cold-winter period (November 2018) the modeled data follow the measured data trend of lower ^{137}Cs activity concentration while during the warm-summer period (July 2019) the modeled data follow the measured data trend of higher ^{137}Cs activity concentration (Figures 6, 7). The difference in the concentrations during the different seasons can be attributed to the circulation of NAS and Black Sea water where during the winter the Black Sea water moves to the North in Samothraki and then moves towards Thermaikos Gulf while during the summer the Black Sea water passes right in between the Islands of Lemnos and Imbros [11, 12]. Overall it seems that the parameter of salinity is capable to predict the ^{137}Cs in the area of Lemnos.

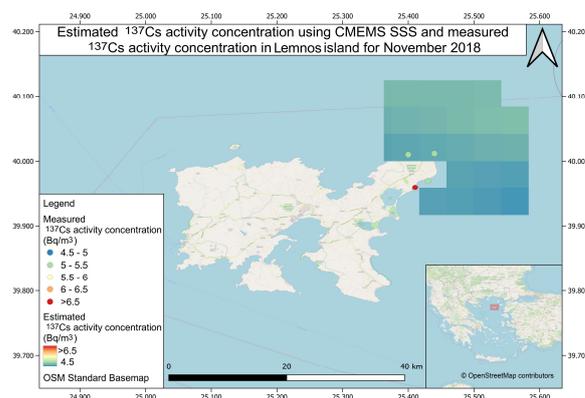


Figure 6. Map of ^{137}Cs activity concentration in Lemnos Island for November 2018. Colored points correspond to measured ^{137}Cs activity concentration values. Pixels correspond to estimated ^{137}Cs activity concentration values issued from the linear model using CMEMS SSS. Basemap from OpenStreetMap (OSM) contributors [15]

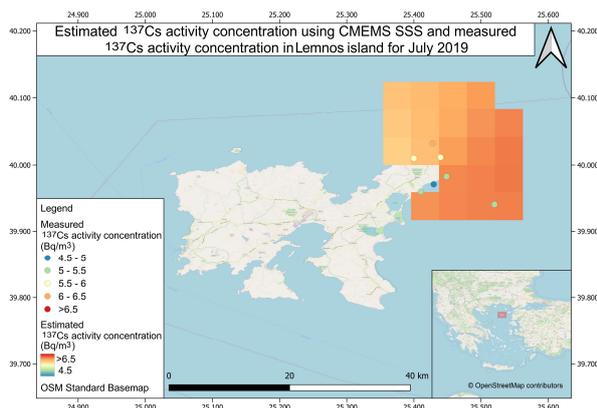


Figure 7. Map of ^{137}Cs activity concentration in Lemnos Island for July 2019. Colored points correspond to measured ^{137}Cs activity concentration values. Pixels correspond to estimated ^{137}Cs activity concentration values issued from the linear model using CMEMS SSS. Basemap from OpenStreetMap (OSM) contributors [15]

4.3. Risk assessment in pelagic fish

The dose rates for pelagic fish seem to follow the trends and distribution of ^{137}Cs activity concentration in the area of Lemnos Island (Figures 8, 9).

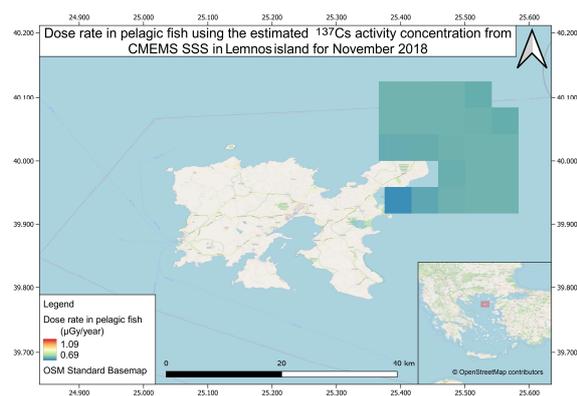


Figure 8. Dose rates for pelagic fish from estimated ^{137}Cs activity concentrations in Lemnos island for November 2018. Basemap from OpenStreetMap (OSM) contributors [15]

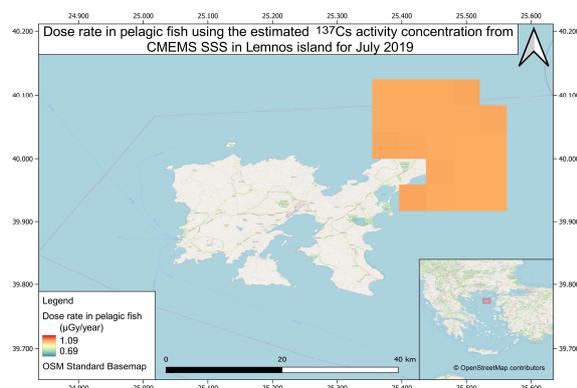


Figure 9. Dose rates for pelagic fish from estimated ^{137}Cs activity concentrations in Lemnos island for July 2019. Basemap from OpenStreetMap (OSM) contributors [15]

Furthermore, the dose rate values also present seasonality, with lower values during the winter and higher values during the summer.

This can easily be explained by the fact that NAS circulation and Black Sea water movements affect the ^{137}Cs activity concentration in the area that is the only contributor to the dose rate (by ^{137}Cs) in the studied organisms in this region [11, 12].

The dose rate in the studied organisms (pelagic fish) that derive by the exposure solely to ^{137}Cs range from 0.7 to 1.1 $\mu\text{Gy}/\text{y}$. As it can be seen, these values are far below the intervention levels (indicatively: 10 $\mu\text{Gy}/\text{h}$), indicating low impact due to ^{137}Cs exposure. However, the integrated risk assessment should normally take into account the exposure to all radionuclide.

5. CONCLUSIONS

In this study the relation of ^{137}Cs and salinity was investigated for the creation of a model that will allow for the estimation of ^{137}Cs , with the purpose of an innovative combination of ERICA Tool and QGIS for risk assessment. The model that was created using CMEMS SSS is able for the estimation-modelling of seawater ^{137}Cs activity concentration in the Lemnos island area. This model also appears able to be used during all seasonal periods as in the area of Lemnos the ^{137}Cs follows the salinity trends. The dose rates to pelagic fish follow the ^{137}Cs activity concentration both temporally and spatially. Moreover they are below than the intervention levels, indicating low impact to pelagic fish due to the exposure to ^{137}Cs .

The use of ERICA Tool and QGIS for risk assessment is innovative and can be used in the future for different radioecological applications. Future work will include the optimization of the model, the creation of an online platform for radiological risk assessment and the study and application of the methodology and tools in other areas.

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