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Towards a sediment reference material database for the Danube River

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Abstract The second largest river in Europe, which has the richest biodiversity within its delta, does not have a sediment reference material database yet. Although there is a plethora of data available, spanning decades and large areas of interest, there is no common ground for data interpretation. The purpose of the database we are starting is to compile the already existing results and our own output from various sources on one hand, and to provide a standardized framework in terms of methodology and uncertainty budget evaluation. This paper briefly presents the procedure proposed for sample treatment and analysis, and focuses on the radionuclide map generated for data visualization. Among the software solutions available, we chose the ones capable of providing clear results for the data integration process in order to obtain a simple, still powerful tool for radionuclide concentration assay, including dynamics. The results are promising and adapted to this need for the Danube River, opening the way to generating a fully documented map for all the natural and anthropogenic radionuclides of interest. The purpose of the present work is not only to track the evolution of nuclide concentration, but also to become able to make predictions.

Keywords environmental radioactivity, gamma spectrometry, Danube River, sediment database, nuclide map

INTRODUCTION

Since the beginning of the Second Industrial Revolution, the pollution due to industrialization has had a tangible impact on air, water and soil quality, changing their composition by introducing TENORM into the natural cycle. Radionuclides dispersed into the atmosphere, through dry/wet deposition, end up on the surface of the earth, which through complex processes of sedimentation, layers of soil contaminated by human activities are produced. Despite the fact that the Danube is the second largest river in Europe and has the richest biodiversity within its delta, a reference material database for sediment is yet to be developed for this area. Although there is a plethora of data available spanning decades of environmental radioactivity research from before the effects of the fallout originated from the nuclear tests in the 60's or the Chernobyl NPP accident to the present day, and covering large areas of interest, from the Danube Delta to the Upper Danube, there is no common ground for data interpretation, reports generally being submitted independently of previous studies.

The purpose of the database we are starting is to compile the already existing results and our own data from various sources on one hand, and to provide a standardized framework in terms of methodology and uncertainty budget evaluation, ideally based on spectra collected with Germanium detectors [1-3]. Using the principles of ^{210}Pb sedimentation (part of Uranium series, therefore naturally occurring and constantly produced), it is possible to identify the dates on which the radionuclides dispersed onto the ground surface, giving us a glimpse of the magnitude of anthropogenic effects on the ecosystem. The moment of conducting the research is relevant because of the limited time frame in which ^{210}Pb can be used for dating (five or six half lives for keeping the relative uncertainty in reasonable limits, meaning some 120 years) in order to encompass a significant, if not the entirety, of

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the time frame of interest. By corroborating data from gamma spectrometry analysis of soil samples extracted from the Danube Delta, with information provided by geological and historical studies of the locations of interest and various other methods for analysis, a comprehensive image of the phenomenon is achieved. Dedicated tools are used to generate a database for specific activities, mapping their concentrations; in order to enhance the exhibition of the dynamics involved in the sedimentation and dispersion processes, several other instruments are used.

The purpose of this project is to develop a system for storing data on radionuclide concentrations and evolution, for mapping and analyzing the dynamics involved, in order to become available to trace origins and also to make predictions on the distribution of radionuclides along the course of the Danube River.

MATERIALS AND METHODS

Sampling points are chosen in order to be representative for the related areas, collected and prepared according to IAEA recommendations [4]; this is valid for all the new lots to be analyzed and introduced in the database to be built. The starting datasets however were provided by various laboratories, mainly from the bordering or passing through countries of the Danube River, and the purpose is to set the results on common ground, as several correction factors are required, the most important being related to the self-attenuation, measurement geometry and real coincidence summing. For those correction factors we use GESPECOR [3], which proved to be robust while versatile enough to carry out the job and provide very accurate results for a multitude of setups. Measurements must be carried out with high efficiency Hyper-Pure Germanium detectors in order to achieve good statistics in a reasonable lapse of time, while providing the necessary fine energy resolution. As many laboratories will be involved, intercomparisons will play a crucial role.

The map is generated using the browser software program Kepler.gl, a versatile open-source geospatial analysis tool for large-scale data sets, in which you can visualize a large amount of multi-layered location data. Filters can be applied to fields of the database in order to display specific information required by the user, such as, but not limited to, sampling dates, radionuclides of interest, ranges of activity concentrations. To create a map, a Comma Separated Values (CSV) file or a JSON file (and its derivatives) is required. The file includes a minimum of three parameters for each data point: latitude, longitude and activity concentration, with the possibility of adding more information depending on the number of the fields that the database possesses which can be toggled as displayable on the data points by moving the cursor on top of them, as displayed in Fig. 1.

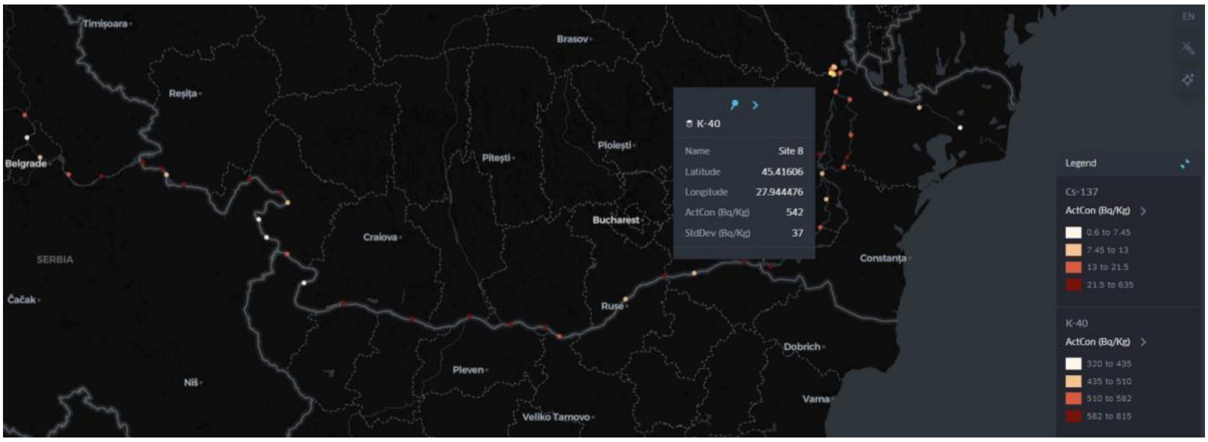

Figure 1. *Map fragment with pop-up textbox displaying information about the selected data point*

A wide range of customizations is available, providing the user the option to select shape and size of data points, color gradient for expressing the ranges of activity concentration relative to the minimum and maximum values stored in the database, with a flexible color palette. Moreover, by adding a filter based on a time-related field, a playback window will appear on the bottom of the map, resulting in the capability for the user to not only select a certain date, but also roll an animation in which the activity concentration fluctuations of the radioelements in time are showcased.

RESULTS AND DISCUSSION

For this application, the data was compiled into a CSV file from several scientific papers dedicated to analyzing the presence of both natural and anthropogenic radionuclides such as ^{40}K, ^{137}Cs, ^{212}Pb, ^{214}Pb, ^{214}Bi, ^{226}Ra, ^{228}Ac, ^{232}Th and ^{238}U in samples collected from sites along the banks of the Danube River. Each point of the map has eight fields allocated in the database, containing the name of the site in which the sample was extracted, the latitude and longitude to accurately identify the geospatial position of the sampling site, the activity concentration and uncertainty of the identified radionuclide, the Digital Object Identifier (DOI) of the article to ensure that the user can access further information regarding the conditions in which the measurements were taken, the date of the sampling and the spectra acquisition time.

The output is the spatial distribution of the radionuclides of interest on a two-dimensional map, each layer representing a radioelement. An example of such is shown in Fig. 1 for ^{137}Cs, compiling data from the first 83 sites by the banks of the Lower Danube River passing through Romania, Bulgaria, Serbia and Croatia. There have been a few numbers of authors which managed to gather and analyze enough samples in order to map the activity of different radionuclides, each in their own regions of interest [5-8]. This approach to data visualization is novel to this geographical area and brings together results from studies on ^{137}Cs concentration in soil samples from 1990 to 2023, while also highlighting the long-lasting impact of the Chernobyl NPP accident on the radiochemical composition of the sediments in the proximity of the Danube River. The number of sites is growing every week, but the goal is to gather cores from all those spots and also new ones and to analyze them as quickly as possible, for ^{210}Pb dating of the layers is running out of time for the early industrialization period, as stated in the introduction. This is a project to be triggered for the European Geological Union General Assembly in 2025.

One thing to be clearly set is the uncertainty evaluation procedure, which must be identical to all the laboratories involved. Uncertainty in gamma ray spectrometry is very well defined [9], but must be treated with great care, for any deviation can produce severe damage to both the inference and the prediction making tool to be developed. Ideally, without having a strict standardization, all laboratories involved should perform their measurements in very similar, if not identical conditions: from detection geometry to sample containers, everything is subject to a new correction factor when it comes to compare a setup to another.

Detector choice is obviously very important, as apart from the high volume semiconductor ones, it is very difficult to take advantage of both high efficiency and resolution. Bias in the data can be avoided by intercomparisons which point out differences and limitations; complementary methods for analysis are always a plus, including for calibration purposes, but most of all, one must consider robust Monte Carlo simulations in order to obtain the proper correction factors and improve analysis accuracy.

CONCLUSIONS

The results are promising and adapted to this need for the Danube River. Data compilation, such as the map showcased in Fig. 1, provides a crucial, wider perspective on the origins and dispersion of certain radioactive pollutants in the natural cycle, while also driving the need to reconcile different methodologies and uncertainty budget evaluations to obtain the most accurate interpretation of the information retrieved from the aforementioned studies. This opens the way to generating a fully documented map for all the natural and anthropogenic radionuclides of interest. Future developments of this project aim to further enlarge the radionuclide database by providing data spanning the entire course of the Danube, with a substantial amount of data in the vicinity of large human settlements such as Vienna, Budapest and Belgrade for an in-depth radiochemical analysis of the influence exerted by the human activities upon the environment. The data in question will be provided by studies on environmental radioactivity done by researchers across the last several decades and data obtained from gamma analysis carried out by our team on samples collected by the various contributors, mainly national institutes for research and development involved in ecological studies or connex activities.

Furthermore, the database will have a number of new fields for each entry, such as the depth at which the sample was extracted, paired with the sedimentation rate of ^{210}Pb which will provide a reliable method of dating, and the measured energy of the respective gamma emission peak. These parameters will be subsequently used for activity determinations. Other parameters might be added after an assessment of their relevance. Later iterations of this map aim to include an algorithm which calculates the activity concentration gradient between any 2 sets of GPS coordinates and simulates the dynamics of the radionuclides with the purpose of developing a system capable of prediction making.

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