Application of Solid Phase Extraction for Automated Sequential Separation of Radionuclides from Environmental Samples

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Radionuclides ingested by food consumption contribute significantly to annual radiation exposure. Main contributors to the annual ingestion doses for Croatian population, consuming locally grown foodstuff, were assessed within "RiChFALL" project by Laboratory for Radioecology of Ruđer Bošković Institute in Zagreb, Croatia. The largest contributors are ^{40}K, ^{228}Ra, ^{210}Pb, ^{210}Po and ^{226}Ra. Therefore, the study of long-term environmental behavior of these radionuclides is of great interest. For determination of-activities by alpha or beta detection, complex sample preparation requires numerous steps such as ion exchanges, precipitations, dissolution, etc. Such time-consuming procedures are expensive, waste chemicals and thus limit the number of analyses in a given time. Novel methods based on solid-phase extraction reduce the work load and consumables, but are still tedious and time-consuming for analyst. Such methods are suitable for automation, which enables efficient use of the analyst's time. In-house automated system for sequential radionuclide separation is being developed at Laboratory for Radioecology of Ruđer Bošković Institute in Zagreb, Croatia. It is comprised of three modules. Control module is Raspberry Pi computer running Linux OS that controls other three modules electronically. Operation of this module is fully programmable via Python programming language. Pumping module is comprised of peristaltic pump connected with plastic tubes between two valves with multiple input tubes. One end of tube network is submerged in various solutions of complexing agents and samples, while the other end is connected to column with resin, e.g. silica-gel modified with crown ether or other complexing agent, which binds radionuclides of interest. The targeted radionuclides, along with all nuclides of specific elements, are retained and concentrated by resin. Separation can be achieved because different radionuclides have different binding constants with various complexing agents. Solutions of those complexing agents, varying in concentration and pH, are sequentially pumped through column. Sampling module is motorized holder for tube coming from the column. It moves in three directions above tray with vessels for different fractions and thus collects eluted radionuclides into separate vessels. Further radiometric analyses such as alpha spectrometry and liquid scintillation counting are carried out "off-line". More than one pumping module and columns with different resins can be interconnected enabling complex separations. Described system minimizes analyst's involvement. It requires only loading of the system with digested, liquefied sample and necessary solutions followed by selection of the desired program with right parameters. Different work modes of the automated system are possible, according to the need, of which two are already considered: 1) Batch operation using single resin and multiple samples and 2) coupling of two resins for complex separations from single sample. Preliminary experiments with reference material and environmental samples from "RiChFALL" project resulted in the separation of radionuclides of Pb, Sr and Ra from foodstuff samples with yields of 65-85% using AnaLig® resin, and the separation of Th radionuclides with a yield of 85-95% using TRU resin. In coupled resins experiments no significant decrease of yields was observed if the order of the resins is AnaLig® then TRU. If the order of resins is reversed, the yield of Ra radionuclides drops to 20%, while yield of Th radionuclides stays the same, so we chose AnaLig® as first resin. The developed methods will be implemented within automated system after programming and individual module tuning. Since the system is modular, it can be rearranged and programmed for new or modified analyses as necessary. The developed system will speed up sample preparation, enable preparations of more samples and reduce analyst's time in the laboratory.