Importance of radionuclide speciation studies - Modelling and Analytical Methods

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The chemical structure of an element (oxidation and coordination states, ligands, charge,…) confers to it specific physical and chemical properties. These latter will act upon the entry, transport, storage of such elements within the body and thus upon the expression of the toxicity of a chemical species. Therefore, to improve the description, knowledge and prediction of both toxicity and absorption kinetic of trace elements, the determination of the chemical species and the assessment of their distribution in a sample or in a tissue are both necessary. These overall analyses are named speciation.

Such studies are essential. Indeed, in case of internal contamination by radionuclides (or other metals or pollutants), speciation studies would enable the improvement of biokinetic and dosimetric models and so allow a better prevention and care.

Several approaches can be used to assess the radionuclide speciation, depending on the accuracy demanded and on the studied tissue.

Among these methods, theoretical approach can be distinguished from the experimental one, both being complementary.

Numerous software and data bases exist and allow the speciation of radionuclides within a solution. Unfortunately, most of the compounds listed in the database are electrolytes, meanwhile proteins or enzymes, which are components of biological tissues and potential chelators are not listed. , …Therefore, the theoretical approach can be efficient for speciation in soil, water, environment media but need to be upgraded to run calculations in biological tissues, particularly by addition of affinity constants of the studied metals and the main organic compounds present in the studied medium.

On an other hand, the assessment of speciation can be performed experimentally.

The main experimental difficulty is to isolate and then to characterize a species without altering it.

Indeed, numerous approaches could be described but in most cases they implied a separation procedure (chromatography, centrifugation, filtration) combined with a detection of the radionuclide. This multi-step analysis is likely to modify the initial complex by changing the ionic strength, pH… or simply remove the compound from its initial mixture.

The detection is also a key-step and must discriminate each species of the mixture in order to characterize it. Several techniques often need to be coupled if the two following properties are not combined: the detection of radionuclide at tiny concentrations and the characterization of the complex. Time-Resolved Laser-Induced Fluorescence (TRLIF) and mass spectrometry (Electrospray Ionisation-MS) are the main candidates for such studies because of their ability to perform the whole assessment. However, Induced Coupled Plasma-Mass Spectrometry (ICP-MS) methods can also be set-up (determination up to nanomolar concentration of an element), coupled with the use of the external standard of each potentially formed complex (peak attribution). Moreover in the case of radionuclides analyses (U for instance), liquid scintillation and α-spectrometry can also allow such measurement, if coupled with the use of external standards.

The example of uranium speciation within saliva will be discussed in this poster. Modelling and experiment will be compared in the case of ingestion of drinking water contaminated with uranyle nitrate.