Behavior of natural and anthropogenic RNs in ecosystems: sources, ecodynamics and impacts

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Main sources of radioactivity in the environment are natural radionuclides (RN) which are ubiquitous in all Earth's surface ecosystems, i.e., in air, soils, waters, and living organisms including Humans, as terrestrial RNs (uranium-238, uranium-235, and thorium-232 from geochemical background and their radioactive daughters) or cosmogenic RNs (e.g. carbon-14). However, during last decades, numerous human activities in the fields of civil or military nuclear industries, or medicine, for example, have led to the production of technologically - enhanced natural radioactive materials (TE-NORM) as well as of artificial RNs that have been released or emitted in natural surface media (mainly by nuclear tests fallout and nuclear accidents). Disposal facilities where low-level radioactive wastes are stored -or planned to be stored- also lie in the Earth's "critical zone" which is the near-surface environment where interactions between rock, soil, water, air and living organisms control natural habitats and life-sustaining resources.

RNs are thus a main environmental issue owing to the increasing variety of RNs in aquatic and terrestrial biotopes, their numerous pathways of introduction or exposure, their potential radio-/chemical toxicity, and the complexity of the bio-physicochemical mechanisms that control their fate and eco-toxicity. For example, soils that contain RNs - even at ultra-trace level concentrations- are potentially the long-term sources for increased transfers to surface waters and to biosphere of cocktails of potentially toxic RNs, which may undergo a bio-magnification along trophic chains and result in eco-physiological damages for living organisms within the chains. Understanding and predicting the transfers / the ecodynamics of the RNs' cocktails, assessing their direct or indirect eco-toxicological impacts and reducing them is thus mandatory and addresses major societal challenges of the 21st century: reduction of anthropogenic risks, sustainability of resources, and preservation of ecosystem's and Human's health.

When a RN is released into a soil-solution system, its transfers, bio-availability, and ecotoxicity is controlled by its chemical speciation (chemical states and forms), which depends on its ability to interact and to form stable chemical species with components within the aqueous, (nano-)mineral and (micro)biological compartments of the ecosystem or at the compartments' interfaces. The concept of chemical speciation is now at the heart of the studies in environmental chemistry, where the priority is no longer to only measure concentrations of RNs but also to develop predictive speciation models allowing to apprehend transfers -and thereby eco-toxicity- of RNs in ecosystems. In order to develop these models, it is of crucial importance to make direct determinations of the speciation of RNs existing at (ultra)trace levels in complex systems and to elucidate the relations existing between speciation, mobility and transfers to trophic chains (bioavailability). To this regard, challenging and emerging research topics deal with elucidating the effects of mineralsolution interfaces, microorganisms, and complex natural organic matter. Meeting these challenges requires the use of powerful and advanced spectrometric, spectroscopic, and analytical techniques in order to carry out molecular-level investigations of the speciation of RNs in complex systems, as well as to quantify transfers at the ecosystem scale of trace RNs by analysis of environmental and biological matrices.

The first part of the lecture will be dedicated to an introduction on the chemistry and functionalities of the soil, and to water-soil interactions.

In the second part, after a brief review of main sources of RNs in the surface environment, we will discuss the recent advances made in the field of the environmental speciation of a selection of RNs and their application to RN behavior in water-soil systems or to safety assessment of low-level waste repositories. Emphasis will be made on the use of advanced spectrometric and spectroscopic techniques.

The lecture will finally highlight briefly new perspectives of innovative transversal approaches developed at the interface between chemistry and biology to validate bio-indicators of the (chemical) eco-toxicological impacts of cocktails of trace RNs.

Some review papers:

- 1. Sparks, D. L. (2003). Environmental Soil Chemistry: an Overview. In. Environmental Soil Chemistry, Sparks (Ed.), Second Edition, 1-42.
- 2. Maher, K., Bargar, J. R., Brown, G. E. (2013). Environmental Speciation of Actinides. Inorg. Chem., 52, 7, 3510-3532.
- 3. Skipperud, L., Salbu, B. (2018). Radionuclides: Sources, Speciation, Transfer and Impacts in the Aquatic and Terrestrial Environments. Encyclopedia of the Anthropocene, 5, 195-206.