Present Research on the Environmental Radioactivity and Its Perspective in Japan

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Understanding of environmental radioactivity has progressed by investigation of undesirable events: nuclear explosion tests and nuclear accidents. Now the importance of the research on environmental radioactivity has realized widely because of an increasing use of nuclear energy. One of the tops of the current research is construction of models, which explain and predict behavior of radionuclides released to the environment. Parameters of mathematical equations used in the model require reliable values, which must be determined by environmental analysis and laboratory experiments. Development of analytical instruments and techniques would make possible to obtain more reliable values of the parameters.

Historical Research Current of Environmental Radioactivity

Our knowledge about the environmental radioactivity has gathered during the past century. The time course of the research on the environmental radioactivity would be divided into three periods as shown in Figure 1. The first period is the dawn of radiochemistry starting with the discovery of radioactivity, the second period is the worldwide contamination by nuclear explosion tests in 1950's and early 1960's, and the last period is the contamination by nuclear accidents coupled with an increasing peaceful use of nuclear energy.

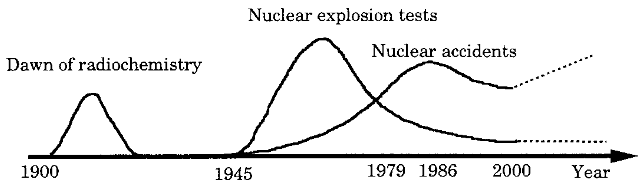

Figure 1. Historical research current of environmental radioactivity.

The discovery of radioactivity by Becquerel in 1896 was carried out by investigation of the possibility that the fluorescence of uranyl compounds. Marie Curie and Pierre Curie showed that all uranium and thorium compounds have radioactive nature independent of chemical composition and found that some of the minerals were much more strongly radioactive than pure uranium itself. This observation led to the discovery of the new elements polonium and radium in 1898. The extensive search of new elements in natural minerals, from the end of the former century to the beginning of this century, is the dawn of the environmental radioactivity. Many new stable and radioactive elements were discovered with great efforts; researchers isolated unknown substances from various kinds of environment materials under poor experimental condition. M. Curie and P. Curie treated a huge amount of rock and isolated polonium and radium by careful purification of pitchblende; it would be the initiation of radiochemical analysis. However, it had been believed that elements are unchangeable forever until E. Rutherford and F. Soddy concluded with careful consideration for the reported observations that radioactivity was due to changes within the atoms themselves. They proposed an epoch-making idea that transformation of the atoms of the original elements to new elements. The investigations done in those days are closely related to the environmental radioactivity.

The first bomb was tested successfully in 1945 and was the signal of the following nuclear explosive races. Many nuclear explosion tests were carried out in 1950's and in the early 1960's, especially those done at open air released large amounts and various kinds of artificial radionuclides into the environment. The artificial radionuclides released into the environment, however, have brought us tremendous increase in understanding of environmental radioactivity. The environmental behavior of the man made radionuclides was extensively examined in the atmosphere, hydrosphere, and biosphere because these radionuclides were the environmental tracer introduced as worldwide scale. For example movement, mixing and diffusion process of water masses in the ocean were studied with helps of tritium, carbon-14 and other radionuclides. Biological accumulation through food webs was well understood by analysis of marine organisms at different levels in the food chain. Analysis of transuranium nuclides, such as plutonium, americium, neptunium, gave us an understanding of chemical, physical and biological behaviors of these nuclides in the environment which did not exist before 1945.

Nuclear accidents at nuclear facilities related to nuclear bomb production have taken place from the beginning of the development. However, most of the accidents were hidden for a long time as military secrets. The atomic weapons complex called "Mayak" at Chelyabinsk province of Russia was closed to people until 1992 in which at least two very large nuclear accidents occurred in 1957 and 1967 which resulted in severe environmental contamination. We experienced large nuclear accidents related to peaceful use of nuclear energy: the Three Mile Island accident in 1979 and the Chernobyl accident in 1986. As an increasing demand of nuclear power generation, the potential for nuclear accidents at nuclear power stations as well as related facilities will be still high in the future. Such accident influences a lot of people because most of the nuclear power stations are constructed near populated areas. In the case of the Chernobyl accident people have been evacuated from the contaminated area to the safety area. The contamination spreads out to the forested area; thus, the behavior of radionuclides in forest ecosystem is the major concern of researchers after the accident. The knowledge about the behavior of radionuclides in the forest ecosystem has accumulated significantly for these ten years; however, it would takes time to understand the whole picture.

The research on environmental radioactivity has progressed in connection with nuclear explosion tests and nuclear accidents, those are undesirable events for the development of

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nuclear energy, and this situation may still continue in near future as the JCO accident recently occurred at Tokaimura, Japan.

Present Research Current of Environmental Radioactivity

Present direction of the study on the environmental radioactivity can be evaluated by screening of scientific programs in recently held symposia. Domestic and international symposia, which included issues on the environmental radioactivity, were held in Japan every year that a total number of eight symposia except regular annual meetings were opened for these two years. The number of the recently opened symposia is considered to be not small, suggesting active research circumstances for environmental radioactivity in Japan. The scientific research contents in the symposia distributed in a wide range from natural radionuclides to artificial radionuclides including nuclear explosion tests and nuclear accidents. A remarkable feature extracted from the recently opened symposia is that an increasing number of presentation concerning models. Prediction of environmental behavior of materials using a proper mathematical model is sometimes required in the field of environmental sciences because of difficulty in doing experiments that reproduce the real environment.

The symposium of "Improvement of Environmental Transfer Models and Parameters" held in Tokyo in 1996 is a good representative of recent research trend for modeling of environmental radioactivity.[1] The sessions arranged in the symposium are listed in Table 1, which well documents the present situation of the model research. The sessions consisted of environmental analysis, model construction for different environmental compartments and interaction between different compartments. The behavior of radionuclides is expressed using mathematical equations in the model and parameters in the equation determine and control results. To construct a good mathematical model, understanding of each environmental process of radionuclide is necessary and also a reliable value of the parameter must be given as shown in Figure 2. Construction of models is impossible unless matching of environmental processes and values of parameters. The values of parameters in the mathematical equations are only obtainable by environmental analysis or laboratory experiments. However, the values thus obtained would have some uncertainties. Even if the laboratory experiments were carried out at a variety of conditions, it does not reproduce the real environment. The values deduced form the environmental analysis are originated from unintended events such as accidents that were uncontrolled or unexpected release; it was not designed for determination of parameter values of the specific environmental processes.

The number of parameters, which should be included in the equation, depends on the purpose and preciseness of model. For a simple model a few number of the parameter would be used. To increase the preciseness of the model, the number of parameter inevitably increases. Chemical, physical and biological processes involved in each radionuclide in the environment are numerous, and an interaction between processes is complicated and is changeable by place, season, climate, etc. It, therefore, seems to be almost impossible to make a perfect model that represent real environment; there is some limitation to evaluate the behavior of radionuclides by model. In spite of the circumstance mentioned above, it would be true that using a model is the only possible way for prediction and evaluation of a whole picture of radionuclides after introduction into a very complicated environmental system. The fate of the radionuclides from the Chernobyl accident is still unclear because movement and distribution of radionuclides in a forest ecosystem is a dynamic process; the results obtained from the environmental analysis revel a cross section of the dynamic process at the moment.

Latest Research and Prospect of Environmental Radioactivity

The recent development in modeling owes to an increasing scientific understanding of environmental processes and also to the social requirement for safety assessments of radionuclides that are released accidentally to the environment. Development of analytical instruments and techniques progresses understanding of environmental processes. Inductively coupled plasma mass spectrometry (ICP-MS) enables us to measure very low level concentrations of elements. Long-lived radionuclides such as uranium and thorium are sometimes detectable for water samples without pre-concentration. Technetium, plutonium, neptunium are able to determine by ICP-MS and ^{239}Pu/^{240}Pu ratio is obtained by ICP-MS that is impossible to measure by conventional alpha spectrometry. Microwave induced plasma mass spectrometry (MIP-MS) is under consideration for ^{129}I measurement. However, the detection limits of the instruments are sometimes not enough to

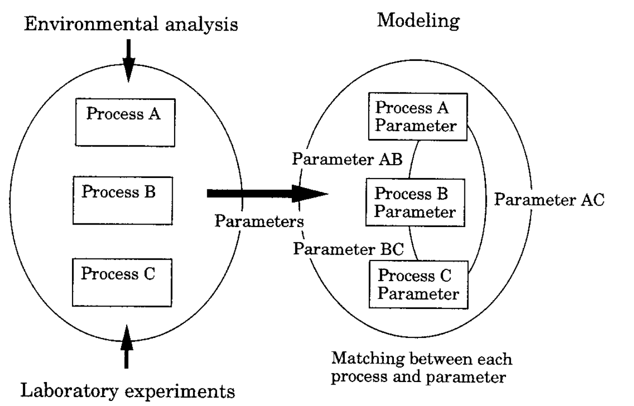

Figure 2. Construction of model based on environmental and laboratory experiments.

TABLE 1: Contents of the International Workshop of "Improvement of Environmental Transfer Models and Parameters"[1] Held in 1996, Japan

Session 1	General topics
Session 2	Transfer parameters
Session 3	Behavior of radionuclides in the terrestrial environment
Session 4	Atmospheric dispersion model
Session 5	Tritium behavior in the terrestrial environment (1)
Session 6	Tritium behavior in the terrestrial environment (2)
Poster session	
A: Transfer of radionuclides from soil to plants	
B: Behavior of radionuclides in soil	
C: Uncertainty of radionuclide behavior in terrestrial environment	
D: Deposition and absorption of radionuclides from atmosphere to plants	

determine radionuclide concentrations involved in each environmental process. Speciation is difficult for some radionuclides by direct measurement, indicating a limitation of reliable parameter values from environmental analysis.

The parameter values obtained from laboratory experiments are often used for the model. Most of the laboratory experiments are designed focusing on a certain environmental process such as change in oxidation state, adsorption on soil, complex formation, etc. Researches related to the interaction between processes are small compared to that of each process. It is almost impossible in Japan to do field experiments using radioactive tracers; such experiments carried out under controlled condition would give us useful information about parameters, including that for interaction of processes. A large experimental facility is under construction at Institute for Environmental Sciences in Rokkasho, Japan, in which quasi field experiments are possible using radioactive tracers under controlling climate.[2] Experimental circumstances to determine more reliable parameters for the model are seemed to be ready soon in Japan.

References

(1) M. J. Frissel, R. M. Brown, and S. Uchida (eds.), *Proceeding of international workshop on Improvement of environmental transfer models and parameters* (Nuclear cross-over research, Japan, 1996).

(2) Y. Ohmomo and N. Sakurai (eds.), *Proceeding of international meeting on Influence of climatic characteristics upon behavior of radioactive elements* (Institute for environmental Science, Japan, 1997).