Radium on soil mineral surfaces: its mobility under environmental conditions
and its role in radon emanation

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I. Research accomplished

The ultimate source of ^{222}Rn to the atmosphere is, of course, ^{226}Ra . Tracking the mobility of radium therefore is part of the story of radon flux assessment. Our study of radium mobility and radon flux measurements has involved virtually all the reservoirs at the Earth's surface. These include soils, groundwaters, coastal waters and the atmosphere. Our attempt to understand the mobility of radium involved the study of almost all the radium isotopes (^{226}Ra , ^{228}Ra , ^{224}Ra) and the parents and daughters of these isotopes.

1. Soil ^{222}Rn flux studies: We have completed an atlas of ^{222}Rn fluxes from soils around the U.S. using ^{210}Pb and ^{226}Ra measurements in soil profiles. Our major effort recently was to calibrate with accuracy the absolute values of ^{226}Ra and ^{210}Pb gamma measurements using a combination of alpha measurements for ^{230}Th and ^{210}Pb on samples chosen to be in secular equilibrium. We have certified the standards available from NIST and Amersham. This procedure was necessary to get absolute ^{222}Rn and ^{210}Pb flux values. Our earlier measurements (Graustein and Turekian, 1990) are now corrected to absolute fluxes and a complete atlas is being prepared this summer.

2. Radium mobility in soil profiles: Using the couples ^{226}Ra - ^{230}Th and ^{228}Ra - ^{232}Th we are able to determine the selective mobilities of the radium isotopes relative to their parents. With the different half lives of ^{226}Ra (1620 years) and ^{228}Ra (5.7 years) we can establish the time scales of the mobility. We have presented this work at meetings and the publication of the results is awaiting final calibration and writing (Graustein et al, 1994).

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3. Mobility of radium isotopes in ground waters: We have established a method for determining the mobility of radium in groundwaters with various chemistries and aquifer properties. Using ^{222}Rn to determine the alpha recoil nuclide flux from the rocks to the ground water the simultaneous measurement of the radium isotopes with different half-lives ($^{226}\text{Ra} = 1620$ years, $^{228}\text{Ra} = 5.7$ years, $^{224}\text{Ra} = 3.4$ days) we obtain the retardation factors for Ra and several other nuclides. These measurements have been made at Yucca Mountain (Nevada Test Site) and Brookhaven National Laboratory as well as elsewhere in Long Island and Connecticut (Krishnaswami et al., 1982; Copenhaver et al., 1992; Copenhaver et al., 1993). The dominant control on the mobility of radium is the redox state of the ground water system. Retardation is high in oxidizing ground waters because of scavenging of radium by manganese and iron oxide surfaces and low in reducing environments because of the absence of these phases.

4. Mobility of radium isotopes in coastal waters: We have studied the distribution and fluxes of ^{226}Ra , ^{228}Ra and ^{224}Ra in Long Island Sound in collaboration with the University of Connecticut Department of Marine Sciences (Turekian et al., 1996; Torgersen et al., 1996). The flux of radium from the sediments was determined as well as the fluxes into and out of the Sound from the East River to the west and the Race to the East. Combining the measurements of these isotopes we could obtain the mean residence time of water in Long Island Sound and the horizontal diffusion coefficients above and below the thermal boundary in the Sound during the summer months.

5. Atmospheric radon and its daughters: One of the ways in which ^{222}Rn is commonly measured in the atmosphere is via its decay products ^{214}Pb and ^{214}Bi . We have explored the ratio ($^{214}\text{Bi}/^{214}\text{Pb}$) to determine to what degree equilibrium had been established for these short-lived members of the decay chain. Other investigators reported wide variations. We found virtually none. In the process we also determined the ratio of ^{214}Pb activity to ^{222}Rn activity and showed ^{214}Pb deficiencies after rain events but equilibrium a short time thereafter. Using ^{214}Pb as a measure of ^{222}Rn in the atmosphere therefore is only possible when rain has not recently occurred. This work was presented at the American Geophysical Union meeting this spring (Turekian et al., 1997) and is in the final stages for publication.

6. Summary: By tracking radium and radon in all of its earth surface reservoirs we have obtained a fuller understanding of the controls on radium migration and radon flux measurements. This pervasive approach to the study of a critical system provides a depth of understanding and yields the potential for other types of studies.

II. Personnel

Part of our goal in our work was to bring a range of scientists together for training and collaboration. The project has had the following people associated with it in its various facets:

Research staff: William C. Graustein, S. Krishnaswami (also PRL, India), Karl K. Turekian.

Collaborators: Henry Shaw (LLNL), Tom Torgersen (U. Connecticut).

Students: Sally Copenhaver, Vaughan C. Turekian. (During the summer, undergraduates from other institutions have participated in the ongoing programs. These are acknowledged in the publications).

These associations have developed a productive web. Among the students, S. Copenhaver is presently at the Lawrence Livermore National Laboratory and V. Turekian is a graduate student at the University of Virginia.

III. References

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Abstracts:

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