

**DOE/ER/14700**

**Aquifer Transport of Th, Ra, and Rn In Solution and Colloids**

**Final Report – 09/15/1996 – 09/14/2000**

**G. J. Wasserburg**

**April 2001**

**Work Performed Under Contract No. DE-FG07-96ER14700**

**For  
U.S. Department of Energy  
Office of Energy Research  
Washington, DC**

**By  
California Institute of Technology  
Pasadena, CA**

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FINAL REPORT

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AQUIFER TRANSPORT OF Th, U, Ra, AND Rn IN SOLUTION AND COLLOIDS

G. J. Wasserburg

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Collaborators: Aude Tricca  
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Brookhaven National Laboratory

Project Number: 55284

DOE DE-FG07-96ER14700

Grant Project Officers: Dr. Nicholas B. Woodward

2 April 2001

3. Executive Summary

The ability to safely store radioactive materials for long times is dependent on our understanding the conditions that mobilize the nuclei. This then requires an understanding of the mechanisms of dissolution and transport in aquifers. This study was directed toward the transport of uranium, thorium and their radioactive daughter products. The purpose was to use naturally occurring radionuclides in understanding the dissolution and transport in groundwater systems without using injected tracers or accidental contaminants. The study involved analyses of groundwaters in and around the Brookhaven National Laboratory (BNL) site and the water supply system. These were the first measurements made of a large number of U-Th series nuclides within an aquifer. A theoretical model of continuous flow was developed considering chemical, physical and geologic properties. This model provides the first self-consistent model of water transport in the vadose zone and the groundwater table with water-rock interactions giving very good insight into the problems of mobilization and precipitation. Clear theoretical predictions on U and Th behavior in groundwaters were derived. The combination of sound theory and good data was successful. Most of the variation in uranium isotopes was shown to be due to the original imprint of near-surface weathering and not to water-rock reactions at depth. It was shown that high Radon contents were not due to micropores in the minerals but reflect thorium precipitation on surfaces during early stages of weathering. Comparison of adjacent but separate aquifers shows that changes in the oxygen content of the water, due most likely to bacterial reactions, can cause the rapid release of large amounts of Th into solution while in oxidized conditions the

retention times are 3,000 years. Thus, subtle changes not easily controlled may alter the long-term stability of such storage systems.

We have been asked to present a review of this work in an invited paper in the hydrology section of the Spring AGU meeting. The work was carried out in a collaboration with three universities and with the support of staff hydrologists at BNL. The two papers that have been published and the AGU abstract are attached as appendices.

#### 4. Research Objectives

We selected a site where it was believed that hydrologic conditions were well understood and where leakage of contaminants had previously occurred. The focus of the project was to find how radioactive nuclei were added to the groundwater system and ultimately to the water supply. Instead of looking for where material was injected and subsequently dispersed, we chose to use the naturally occurring radioactive nuclides of the U and Th series that are present in the soils and rocks through which the water flows. By using this approach and the distinctive chemical properties and mean lifetimes of the nuclides it was, in principle, possible to infer the mobilization and transport mechanisms under natural conditions. One limitation was that actual knowledge of the hydrologic flow field is not nearly as well described as would be needed. This problem remains. The distribution of the nuclides between dissolved, colloidal and particulate matter was determined. Using a series of related nuclei whose production is known, it is possible to deduce the chemical and transport behavior that has occurred. This was done using the isotopes  $^{232}\text{Th}$ ,  $^{234}\text{Th}$ ,  $^{228}\text{Th}$ ,  $^{238}\text{U}$ ,  $^{234}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{224}\text{Ra}$ , and  $^{222}\text{Rn}$ . The data were considered in the context of a theoretical flow model that we developed. This model included near-surface and deep weathering,  $\alpha$ -recoil, exchange with a surface layer on the rocks, “irreversible” precipitation, porosity, water flow rate and distance. It was possible to obtain simple mathematical solutions in closed form. These led to clear predictions of behavior and, with the data, provided a means of checking the model.

5. Methods and Results

The methods and results are described in full detail in publications in the appendix.

6. Relevance, Impact, and Technology Transfer

We consider the relevance to be high. The directors of BNL and the staff hydrologists are being provided with the full report, which is now published.

7. Project Productivity

We accomplished a great deal but did not accomplish as much as I had wanted. We had to eliminate the work on the Edwards aquifer. The fundamental problem is that there are very few talented people with good training who are interested in this field. In particular, the number of qualified U. S. citizens who applied for positions was incredibly low. Unless we are able to get really talented and skilled people into the field, we are in trouble. To a large extent the fault lies within the university programs, their standards and training. The people in environmental engineering and the isotope people really must be brought into working together. The need for sites where the hydrology and flow (e.g., flowlines) are understood remains a problem.

8. Personnel Supported: Aude Tricca, France, Don Porcelli, USA, G. J. Wasserburg, USA; J. Naidu, M. Baskaran.

9. Publications:

Tricca A., Porcelli D., Wasserburg G. J., and Baskaran, M. (1998) Transport of U and Th-series radionuclides in groundwater. 8<sup>th</sup> Vv. M. Goldschmidt Conference, Toulouse, France. Mineralogical Magazine 62A, 1543-1544.

Tricca A., Porcelli D., Wasserburg G. J., Baskaran, M., and Naidu J. (1998) Development of a transport model of U-Th decay series nuclides in groundwater and its application to a sandy aquifer. *Eos, Trans. Amer. Geophys. U.*, 79, F425.

Tricca A., Porcelli D., Wasserburg G. J., and Baskaran, M. (2000) Transport of U and Th-series radionuclides in a sandy aquifer. DOE Conference, Atlanta, GA, April 2000.

Tricca A., Porcelli D., Wasserburg G. J., and Baskaran, M. (2001) Transport of U and Th-series radionuclides in a sandy aquifer. In press, *Geochim. Cosmochim. Acta*.

10. Interactions

Presentations have been made at three international meetings and one conference at Harvard University. Collaboration was with University of Texas, Galveston and the Department of Geology, Wayne State University (M. Baskaran). Support from the hydrological staff (J. Naidu, D. Paquette, and R. Lagatolla) of BNL was obtained.

11. Transitions: No examples.

12. Patents: None

13. Future work: The theoretical approach resulting from this project now requires testing in other areas. In particular hydrologic systems with low flow rates and/or long flow paths should be the target. The question of “the age” of groundwaters and the extent of water-rock interaction must be generalized. The most difficult problem is to find aquifers that are sufficiently well understood. Inferences based on inadequate knowledge of an aquifer turn into using the chemistry to infer aquifer flow. This is useful but does not expose the first principles that we are seeking. The other major issue is related to

Radon. We must now establish whether the inference that early weathering coats rocks with  $^{232}\text{Th}$  and  $^{230}\text{Th}$  and thus is the source of  $^{222}\text{Rn}$  must be directly tested.

14. Literature Cited: None in this report. The publications listed in the Section 9 record the literature used in the study as a whole.

15. Feedback: See Section 13.