Reports

Migration of Ruthenium-106 in a Nevada Test Site Aquifer: Discrepancy Between Field and Laboratory Results

Abstract. *Ruthenium-106 has been observed to migrate in ground water at about the same velocity as tritium from the site of an underground nuclear explosion to a pumped satellite well 91 meters distant. This finding contradicts the prediction, based on laboratory batch sorption measurements, that ruthenium-106 should migrate at a much lower rate than tritium. To predict migration of radionuclides in ground water, more relevant laboratory measurements are required.*

To assess the potential for migration of radionuclides deposited in aquifers at the Nevada Test Site by underground nuclear explosions, a program was begun in 1973 by the Department of Energy's Nevada Operations Office to study the interaction of nuclear explosion debris and ground water (*1*). Results from this program also elucidate what might happen in a flooded nuclear waste repository. We measured the migration rate of ruthenium-106 in the field and found a significant discrepancy between our results and the rate calculated from laboratory batch sorption measurements.

The radionuclides we measured were produced in the Cambric nuclear test in May 1965. The device was detonated at a depth of 294 m in tuffaceous alluvium. To determine the concentration of radioactivity in ground water in direct contact with nuclear explosion debris, water was sampled from a well that intersected the explosion cavity (Fig. 1). Tritium and strontium-90 were the only radionuclides whose concentrations were above the maximum permitted for unrestricted public use of drinking water, even though the water had been in direct contact with nuclear debris for 10 years (*2*).

During April 1974 a satellite well was drilled 91 m south of the explosion point to study migration rates of the radionuclides in the cavity water. Pumping from the satellite well induced a gradient between the explosion point and the well. No radioactivity was observed in the satellite well until 1.44×10^6 m^3 of water had been pumped over a period of more than 2 years (*3*). With further pumping, the ^{3}H concentration continued to increase in the satellite well and decreased 50-fold in the cavity well (*3*).

After establishing that cavity water had been drawn to the satellite well, we attempted to measure radionuclides other than ^{3}H. From calculations based on the known relative concentrations of radionuclides and the dilution factor measured for ^{3}H, we concluded that the activities of other explosion-produced radionuclides would be below the limits of analytical detection in standard water samples, even without sorption.

To solve this problem, we collected 200-liter water samples and evaporated them in the laboratory. The radioactivity of the resultant salts, whose mass was about 150 g per sample, was measured with an ultralow-background Ge(Li) Compton suppressed gamma-ray spectrometer (*4*). To attain even greater sensitivity, a field distillation apparatus was designed and built to evaporate up to 1000 liters of water at the wellhead. To date, three samples from 700 to 860 liters have been evaporated in the field and the resultant salts measured for radioactivity.

Nearly the same levels of the natural radioactive species ^{40}K, ^{235}U, and ^{238}U were observed in all samples (Table 1). Ruthenium-106 was also observed in all samples. The ^{106}Ru activity tended to increase with time, although the errors due to counting statistics were generally large. The ^{106}Ru/^{3}H ratios were nearly the same for all water samples from both wells. This suggests that ^{3}H and ^{106}Ru traveled at the same rate from the explosion site to the satellite well.

Contamination was ruled out at our laboratory by an analysis of two unidentified specimens prepared from 200-liter samples that had been collected by Los Alamos National Laboratory. One sample had a ^{106}Ru content comparable to that of one of our samples. The other showed no ^{106}Ru but had similar levels of ^{40}K, ^{235}U, and ^{238}U. This sample turned out to be a blank from a Nevada Test Site well which is located 2.25 km from the satellite well and pumps water from the same aquifer at a depth of 230 m. We regard field contamination as unlikely because the ^{106}Ru content approximated the predicted value (*5*), no other explosion-related radionuclides were observed, and the ^{106}Ru/^{3}H ratios of the samples were similar to the values for the cavity well.

The calculated ^{106}Ru/^{3}H ratio for the amounts produced by the nuclear explosion is about 100 times higher than that observed in water samples from the cavi-

Table 1. Radioactivity in salts from evaporated large-volume water samples from the satellite well at the Cambric site. Data were recomputed to 15 years after the 1965 explosion so that they are closer to current radioactivity levels. For each value (picocuries per liter), the stated error (in parentheses, expressed as a percent) is based only on the counting statistics. We have been collecting 200-liter samples from the satellite well since April 1977. Some have not been evaporated, and a few (not reported) were contaminated.

Radionuclide	Date of sample and volume (liters)						
	Blank* (200)	11/29/78* (200)	2/7/79 (200)	3/14/79 (200)	8/29/79 (710)	4/17/80† (740)	7/23/80 (860)
^{40}K	8.6 (2)	10 (2)	7.2 (3)	7.7 (2)	10 (2)	12 (1)	8.6 (1)
^{106}Ru	≤ 0.02	0.04 (45)	0.04 (40)	0.04 (30)	0.07 (7)	0.08 (45)	0.08 (21)
^{235}U	0.07 (6)	0.09 (6)	0.05 (7)	0.08 (5)	0.04 (4)	0.12 (7)	0.09 (2)
^{238}U	1.9 (5)	2.5 (5)	1.0 (35)	1.6 (26)	0.90 (6)	3.0 (16)	2.2 (4)

*Supplied by Los Alamos National Laboratory. The blank was taken from Nevada Test Site well 5B, located 2.25 km south of the Cambric site. At the time of the analysis we did not know that this was a blank sample. Well 5B is the water supply well for Mercury, Nevada, but draws from the same aquifer as the satellite well. †This sample was tested with a Ge(Li) gamma-ray spectrometer different from that used for the other samples. Since this spectrometer had a higher background, the ^{106}Ru was more difficult to detect. This is shown by the high counting statistics errors for ^{106}Ru, ^{235}U, and ^{238}U.

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ty well or the satellite well. Since ^3H exchanges rapidly with the hydrogen in water, most of the ^3H produced in the explosion is probably in the ground water. However, since ^{106}Ru can become immobilized in the melt glass (*1*) or be unavailable due to sorption or precipitation, only about 1 percent of the total produced became a mobile species in the ground water.

Some 120 pCi of ^{106}Ru per liter had been found in the cavity well water before pumping began at the satellite well, but after 4 years of pumping a 400-liter sample from the cavity well contained no ^{106}Ru above the detection limit (0.01 pCi/liter). This indicates that the mobile species was swept from the cavity and not replenished by leaching of melt glass or by reversible reaction from a sorbed or precipitated form.

Ruthenium is reported to have equilibrium sorption coefficient (K_d) values in ground water ranging from 10 to 8000 (*1*, *6*–*8*), based on batch sorption techniques. Even a K_d as low as 10 for this rock results in a retardation factor of 64. That is, the nuclide would move at a rate 1/64 that of the water and by July 1980 would have moved only 2.7 m. However, the lowest K_d for ^{106}Ru measured (with rock and water from this site) is 976 (*6*), which would result in only 3 cm of movement. Only nuclides with a $K_d \leq 0.3$ would have migrated the 91 m to the satellite well by July 1980.

The laboratory batch sorption method gives only the average K_d for all chemical forms of a radionuclide. The large range in reported K_d values for ^{106}Ru could result from a differing proportion of chemical forms in each experiment. These forms could have a wide variation in individual sorption properties, including one or more with an effective K_d of zero.

Ames and Rai (*8*) report that ruthenium exists in solution only as complex ions. On the basis of simplified *Eh* (oxidation-reduction potential-*p*H diagrams for the range of *Eh* and *p*H values expected for ground water, Brookins (*9*) indicated that three forms could exist, depending on *Eh* conditions and the presence of certain anions. For reducing conditions, solid RuS_2 is a potential stable phase. For more oxidizing conditions, solid RuO_2 is a potential stable phase. At still stronger oxidizing conditions, a stability field exists for the complex ion RuO_4^{2-}.

Laboratory batch sorption experiments on Hanford, Washington, soil produced K_d values from 40 to 752 within the *p*H range of 7 to 9 expected for ground water (*8*). In contrast, field observations near leaking storage tanks or waste pits showed significant mobility for ruthenium, and in one case ruthenium migrated with tritium and technetium (*10*). A review by Onishi *et al.* (*11*) concluded that ruthenium migration in the field has been well documented; on the other hand, laboratory tracer experiments have shown high K_d values for ruthenium.

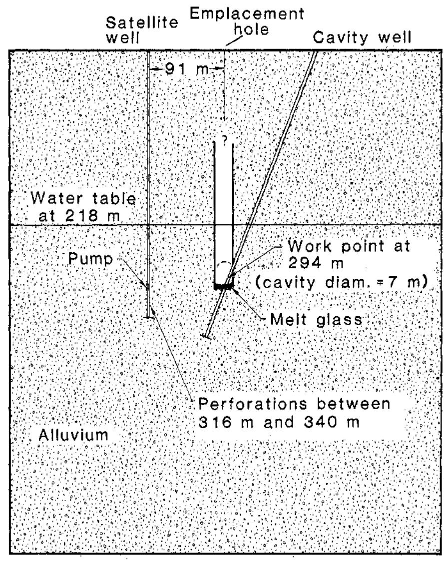

Fig. 1. Cross section of the Cambric experiment site.

The ground-water chemistry of the tuffaceous alluvium aquifer of Frenchman Flat is documented from several wells near the Cambric site (*2*, *6*, *12*). The composition of the satellite well is typical of Frenchman Flat ground water. It has often been assumed that deep aquifers (> 250 m) are reducing since they are isolated from the atmosphere. However, recent studies by Winograd (*13*) showed that many deep aquifers, including Nevada Test Site ground water, are nearly saturated with dissolved oxygen. Wolfsberg (*6*) measured the *Eh* with a platinum electrode at the satellite well and observed mildly oxidizing conditions (+330 mV). Considering the dissolved oxygen content in these waters and the *Eh*-*p*H diagrams for ruthenium (*9*), we would expect some migration of ruthenium, probably as RuO_4^{2-}.

The fact that ^{106}Ru migrated at rates similar to those of ^3H in a tuffaceous alluvium aquifer at the Nevada Test Site does not indicate a hazard from potential ground water migration from nuclear tests to off-site wells or springs. First, the initial concentration of ^{106}Ru in the chimney itself is well below the maximum permissible concentration for drinking water (*2*). Second, the half-life of ^{106}Ru is so short (1.01 years) that the radionuclide would never reach distant wells before it had completely decayed. Third, only ^{106}Ru has been observed to migrate.

The field observations reported here do not invalidate all batch sorption measurements, but K_d values thus obtained should be used with caution and verified with field radionuclide migration studies or more relevant laboratory studies. For elements that usually have a single valence (such as strontium and cesium), the batch K_d values appear to have relevance if sorption isotherms are determined. For multivalent elements, flow system methods rather than batch methods may be required to provide data relevant to field conditions. In addition, an understanding of the chemical speciation of multivalent elements like ruthenium, technetium, and selected actinides is needed in order to predict their behavior in a ground-water environment.

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References and Notes

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Alzheimer's Disease and Senile Dementia: Loss of Neurons in the Basal Forebrain

Abstract. *Recent evidence indicates that the nucleus basalis of Meynert, a distinct population of basal forebrain neurons, is a major source of cholinergic innervation of the cerebral cortex. Postmortem studies have previously demonstrated profound reduction in the presynaptic markers for cholinergic neurons in the cortex of patients with Alzheimer's disease and senile dementia of the Alzheimer's type. The results of this study show that neurons of the nucleus basalis of Meynert undergo a profound (> 75 percent) and selective degeneration in these patients and provide a pathological substrate of the cholinergic deficiency in their brains. Demonstration of selective degeneration of such neurons represents the first documentation of a loss of a transmitter-specific neuronal population in a major disorder of higher cortical function and, as such, points to a critical subcortical lesion in Alzheimer's patients.*

Alzheimer's disease (AD) and senile dementia of the Alzheimer's type (SDAT) are associated with progressive deterioration of memory and cognitive function (*1*). These two disorders, differing in the age of onset and rate of progression, but similar in terms of pathology (*2*), are the most common causes of dementia in middle and late life. Perhaps 10 to 15 percent of the population over 65 years of age suffers from mild to severe dementia; 50 to 60 percent of these patients have SDAT, resulting in a prevalence of over a million affected individuals in the United States (*3*). Neurochemical investigations of patients with AD/SDAT have demonstrated a reduction in presynaptic markers for acetylcholine-utilizing neurons (*4*) in the hippocampus and cerebral cortex. The basis for this cholinergic abnormality is unclear, but recent evidence suggests that the cholinergic neurons in the nucleus basalis of Meynert (nbM) may selectively degenerate in AD/SDAT. The nbM (*5*), located in the substantia innominata (Fig. 1), contains clusters of neurons that can be recognized on the basis of their large size, abundant Nissl material, acetylcholinesterase (AChE) activity, and physiological properties (*5–8*). Choline acetyltransferase (CAT) activity, the best available marker for cholinergic neurons (*9*), is enriched in the substantia innominata, and the distribution of this enzyme parallels the topography of the large neurons in the nbM (*10*). These large neurons in the nbM project directly to the cerebral cortex (*11, 12*); similar neurons, located in the diagonal band of Broca and medial septum, project to the hippocampus (*13*). Moreover, excitotoxic lesions of the ventral pallidum, which, in the rat, contains cells homologous to the neurons of the primate nbM, cause a selective reduction in cholinergic presynaptic markers in cortex (*14*), similar to that described in patients with AD/SDAT (*4*).

These observations directed our attention to the basal forebrain in AD/SDAT, and we recently have shown a 90 percent loss of neurons in the nbM in a patient with a familial form of AD (*15*). We now describe a selective degeneration of neurons in the nbM in five patients with presumed sporadic AD/SDAT. We conclude that the loss of putative cholinergic neurons in the nbM is linked to the presynaptic neurochemical abnormalities in the cortex of patients with AD/SDAT.

The brains of demented patients, obtained from the collection of the Maryland State Medical Examiner's Office, were selected on the basis of three criteria: a history typical of AD/SDAT; the presence of the classical pathology of AD/SDAT including neurofibrillary tangles, senile plaques, and granulovacuolar degeneration; and the availability of paraffin-embedded, Nissl-stained histological sections (15 μm thick) containing the most extensive portion of the nbM, for example, the substantia innominata between the optic tract and anterior commissure. The brains of five adults without evidence of dementia were selected for study on the basis of comparable age and availability of matching histological sections of the ventral forebrain (Table 1). Cells were counted as nbM neurons on the basis of three criteria: cell size (larger than 30 μm); the presence of abundant Nissl substance; and a visible nucleolus. Cell counts (Table 1) were performed by two observers, blind to the diagnosis, using three different assessment methods. Cell loss was judged subjectively, according to a seven-point rating scale with 0 corresponding to complete loss of cells and 6 corresponding to normal cell number. The number of neurons contained within a zone measuring 600 μm by 600 μm was counted in the area of maximum cell density and was expressed as the average number of cells per grid (Table 1); and the total number of neurons within histological sections containing the major (midportion) of the nbM was counted directly. If the nbM's on both sides were available for counting, the average cell count on each side was used.

In each of the patients with AD/SDAT, all three methods of assessment showed severe loss of nbM neurons (Fig. 2 and Table 1). The maximum cell density of neurons was reduced by 73 percent, and the total number of neurons was reduced by 79 percent. In each independent rating scale, there was fourfold difference

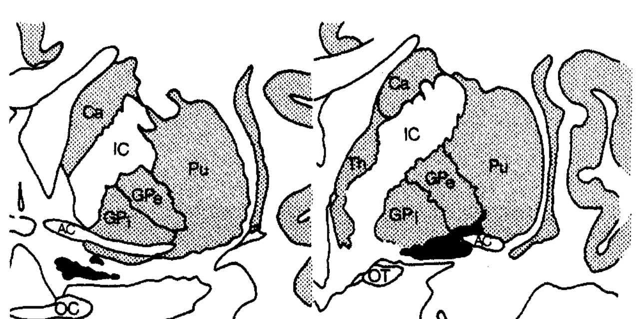

Fig. 1. Drawings of the human forebrain, adapted from (*6*), illustrating the anatomical relationships slightly rostral to and caudal to the forebrain sections available for morphological analysis. The region containing the nbM is shown in black. (A) Basal forebrain at the level of the optic chiasm. (B) Basal forebrain at the level of the infundibulum. Abbreviations: *AC*, anterior commissure; *Ca*, caudate nucleus; *GPi*, internal segment of the globus pallidus; *GPe*, external segment of the globus pallidus; *IC*, internal capsule; *OC*, optic chiasm; *OC*, optic chiasm; *OT*, optic tract; *Pu*, putamen; and *Th*, thalamus.

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