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Mechanism of Plutonium Transport in a Shallow Aquifer in Mortandad Canyon, Los Alamos National Laboratory, New Mexico

RICHARD C. MARTY,*,† DEBORAH BENNETT,‡ AND PHILIP THULLEN‡

5075 North 51st Street, Boulder, Colorado 80301, and Los Alamos National Laboratory, Los Alamos, New Mexico 87545

Potential environmental effects of radioactive isotopes are of great public concern. Fortunately most long-lived isotopes have strong affinities for solids and limited mobility under natural conditions. It recently has been proposed that some isotopes may form colloids that move with groundwater. The detection of plutonium in groundwater some 3400 m down Mortandad Canyon at the Los Alamos National Laboratory is cited widely as an example of such transport. The current work re-examines data from this canyon to evaluate the significance of such transport. ^{239}Pu entering the canyon increased sharply in the early 1980s. Routine monitoring during this period shows that isotopically distinct plutonium appeared in one downgradient well before it appeared in wells closer to the source. If this is ignored, plutonium moved at least twice as fast as groundwater flow and easily outdistanced a tritium peak. Isotopically heavy plutonium arrived simultaneously in surface alluvium and groundwater, and the isotopic composition of plutonium in alluvium and groundwater are identical. Plutonium clearly did not move down-canyon via groundwater. The potential for plutonium movement through groundwater on colloids may be overstated at this and other sites.

Introduction

The Los Alamos National Laboratory was constructed in the early 1940s in northern New Mexico on the site of a former boy's school and served as the U.S. center for design and construction of atomic weapons. The world's first atomic weapons were constructed at the site during World War II, and the facility continued to be used after the war to improve weapon design, construct new weapons, and develop nuclear technologies. Over the years, such activities have produced a variety of environmental problems.

The laboratory lies on the deeply eroded peneplain of the Pajarito Plateau, which slopes eastward from the Jemez Mountains to the Rio Grande Valley. Laboratories, living quarters, and support facilities were constructed on the east–west trending mesa tops of the plateau, and the canyons incised into the plateau were used to dispose of solid and liquid waste. Some of these wastes were contaminated with traces of radioactive and hazardous constituents that are byproducts of nuclear research, and over the years the canyons have received measurable quantities of plutonium together with mixtures of other actinides, fission products, and activation products. Some isotopes of plutonium have long half-lives, high toxicity, and high carcinogenicity, and consequently plutonium released to the canyons has been studied extensively over the years.

Most studies have concluded that plutonium is strongly sorbed by soils and sediments and that actinide contamination moves downslope primarily through erosion and redeposition of the contaminated soils and sediments (*1–3*). Recent workers have suggested, however, that actinides sorbed on fine-grained colloidal-sized materials have moved quickly through shallow groundwater in one of the canyons (Mortandad Canyon) via the mechanism of colloidal transport (*4*).

Under certain conditions, colloidal-sized particles may transport insoluble contaminants through aquifers without significant retardation on aquifer materials. The particles must have diameters smaller than the stationary media, the particles must have neutral or negative charges, the contaminant must be strongly held by the mobile phase, and the migrating colloids must not be removed from the water (*5–7*). Clays, hydrated iron oxides, hydrated aluminum oxides, high molecular weight organic molecules, and actinide hydroxides have all been suggested as possible mobile phases under some conditions (*7–11*). Transport of plutonium in this manner would be significant because it could allow insoluble plutonium to move quickly into drinking water supplies, potentially threatening human health and the environment.

The Los Alamos National Laboratory has collected a large body of data on the geochemistry of groundwater and alluvium in Mortandad Canyon, and much of this data has not been incorporated into earlier studies of the movement of plutonium in the canyon. During the summer of 1994, the available data was reviewed by the authors as part of an independent technical review of the Environmental Restoration Program at the Laboratory commissioned by the laboratory (*12*). The portions of this review which deal with plutonium transport in Mortandad Canyon have been refined and focused and are presented below.

Background

Under unperturbed natural conditions, surface water flow in Mortandad Canyon is ephemeral, and the canyon lacks perennial reaches (*13*). Mortandad Canyon, however, has received non-radioactive wastewater from TA-46 and treated effluent from TA-50 since 1963, and these discharges produce perennial flow in the middle of the canyon (*14*). The water generally evaporates or percolates into the canyon fill within 2 km of the discharge, but spring snowmelt and periodic storms can produce flow 3.4 km or further from the outfall (*15, 16*). Surface water has not flowed off-site since effluent discharge began in the 1960s, and a series of traps were constructed in the early 1970s to prevent any off-site migration of radioactive surface water in the future (*17*).

Alluvium in Mortandad Canyon forms a thin ribbon that thickens down canyon from 0 m in the west to 30 m thick in the east. The alluvium consists of variable quantities of clay, silt, sand, and gravel derived from the underlying Bandelier Tuff (*2*). The Bandelier Tuff is up to 300 m thick, and the uppermost member of this tuff (the Tsherege Member) is relatively impermeable (*13, 18*). This impermeable layer blocks the downward percolation of seasonal runoff and treated wastewater, and water perches in the alluvium creating a small groundwater body (*1*).

* Corresponding author telephone: (208) 525-9358; fax: (208) 525-3364; e-mail: 104363.746@compuserve.com.

† Current address S.M. Stoller Corporation, 447 Park Avenue, Idaho Falls, ID 83402.

‡ Los Alamos National Laboratory.

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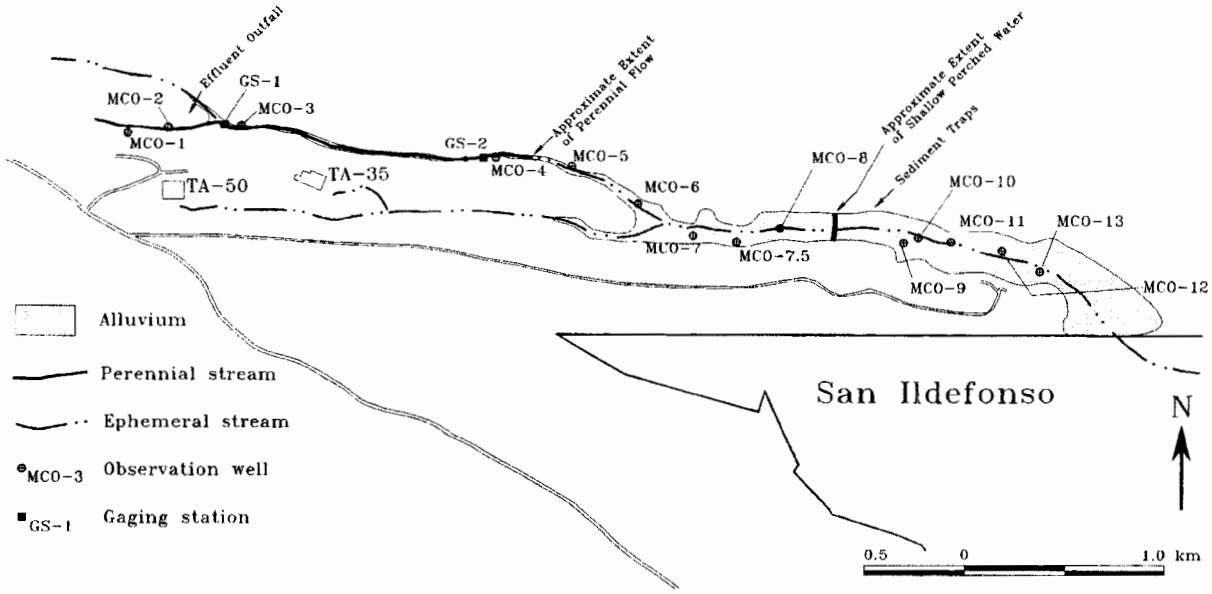

FIGURE 1. Locations of monitoring wells and other features in Mortandad Canyon at the Los Alamos National Laboratory (after ref 36).

The groundwater body is entirely on laboratory property (Figure 1), and its downstream extent is controlled by evapotranspiration and percolation into underlying rocks (*19*). The shallow groundwater occurs in a sand in the upper canyon to the west of monitoring well MCO-5, in a transitional silty clay in the middle canyon between monitoring wells MCO-5 and MCO-6, and in a silty clay in the lower canyon to the east of monitoring well MCO-6. Groundwater flows approximately 18 m/day in the sand, approximately 6 m/day in the transitional zone, and approximately 2 m/day in the silty clay (*20*).

The movement of plutonium down the canyon has been studied in some detail (*1, 2, 4, 14, 19, 21–23*). Plutonium enters the canyon in effluent from TA-50 and is concentrated on alluvium near the outfall. The plutonium has a high affinity for clay- and silt-sized fractions of alluvium, but the more abundant sand-size fraction contains most of the plutonium inventory (*21*). About 14% of the plutonium is associated with silt and clay, which comprise only 2% of the alluvium, but over 85% of the plutonium is associated with sand (*2, 14*).

Plutonium builds up in alluvium near the effluent outfall, but this buildup is limited by erosion and transport of plutonium-bearing alluvium down the canyon during periods of high flow (*21*). Clay and silt are carried in suspended load, and sand and silt are carried as bed load (*24, 25*). While runoff from snow melt during the winter and spring can carry significant quantities of plutonium, most transport occurs during flash floods and other periods of high flow during the summer and fall (*22, 26*).

More than 99% of the total plutonium inventory in the canyon is associated with alluvium in or immediately adjacent to the stream channel (*27*). Stream-bank alluvium up to 1 m on either side of the channel contains approximately the same levels of plutonium as the channel, and plutonium levels can reach a few hundred picocuries per gram locally (*2, 13, 14*). Plutonium is rather uniformly distributed within the alluvium to depths of 30 cm as a result of physical mixing during storm runoff, and elevated plutonium concentrations are found at depths of up to 100 cm in the channel and depths of up to 50 cm in the stream bank (*2, 14*).

The affinity of the alluvium for plutonium results primarily from exchange by clay minerals (*14, 20, 28*). Sorption of plutonium is interpreted to be a surface reaction with clay minerals (*2*), but the reaction is not quickly reversible, and plutonium bound on colloids will not equilibrate with isotopically spiked solutions even over a period of days (*4*). A strong affinity between plutonium and aquifer materials also has been documented in studies of the migration of plutonium in boreholes drilled into the unsaturated Bandelier Tuff beneath the perched water zone in Mortandad Canyon. After 30 years of radioactive releases from the treatment facility, plutonium moved less than 3 m into the tuff (*27*). This is consistent with an apparent soil–water distribution coefficient (K_d) for plutonium of approximately 20 000 L/kg determined for particles trapped on 0.45-μm filters and water passing through the 0.45-μm filter (*4*).

TABLE 1. Well Construction Information[a]

well	completed	top of screen or perforation[b] (m)	base of screen[b] (m)
MCO-3	Mar 1967	0.61	3.7
MCO-4	Oct 1963	1.2	5.8
MCO-5	Oct 1960	6.4	14.0
MCO-6	Mar 1974	8.2	14.3
MCO-7	Oct 1960	11.9	21
MCO-7.5	Apr 1974	10.7	18.3

[a] Compiled from ref 37. [b] All depths are below land surface.

The high affinity of the sediments for plutonium prevents dissolved phase transport of plutonium through groundwater, but plutonium has been detected in groundwater as far as 3.4 km from the effluent outfall. The mechanism of plutonium movement into the monitoring wells has not been unequivocally determined, but variation in the apparent K_d for ^{241}Am on >0.45-μm particles down canyon has been suggested as evidence of colloidal transport of actinides through the groundwater. Apparent ^{241}Am K_d values for particles trapped on 0.45-μm filters and water passing through the 0.45-μm filter varied from approximately 10^4 L/kg at 1700 m from the outfall to approximately 10^2 L/kg at 3400 m from the outfall in samples collected during 1982 and 1983 (*4*). Americium is strongly bound to colloidal materials smaller than 100 000 MW (approximately 0.025 μm), and the difference in apparent K_d resulted from relative enrichment of small, high americium-affinity colloids in the downgradient wells. If storm events and subsequent migration of contamination into monitoring wells are assumed to move all size fractions less than a few microns with similar efficiency, then enrichment would argue that actinide contamination resulted from colloidal transport (*4*).

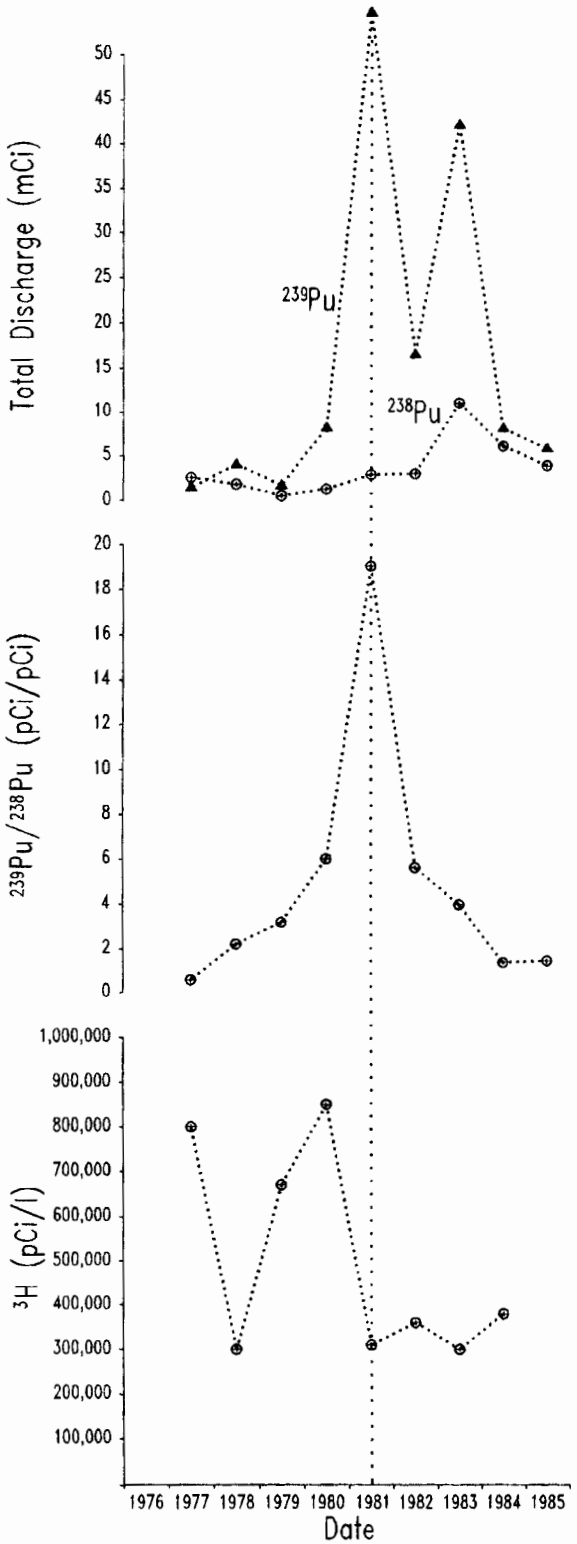

FIGURE 2. Variations in effluent composition through time. The amount of ^{239}Pu and the $^{239}Pu/^{238}Pu$ ratio both peaked in the early 1980s (data from refs 3, 15, 16, and 29−35).

Data

Radioactive constituents are monitored systematically in the various media in the canyon and form an extensive data set (*3, 15, 16, 29−35*). Data on ^{238}Pu, ^{239}Pu, and tritium in effluent, shallow groundwater, and surface alluvium through time are fairly complete, and the compiled data are useful in testing

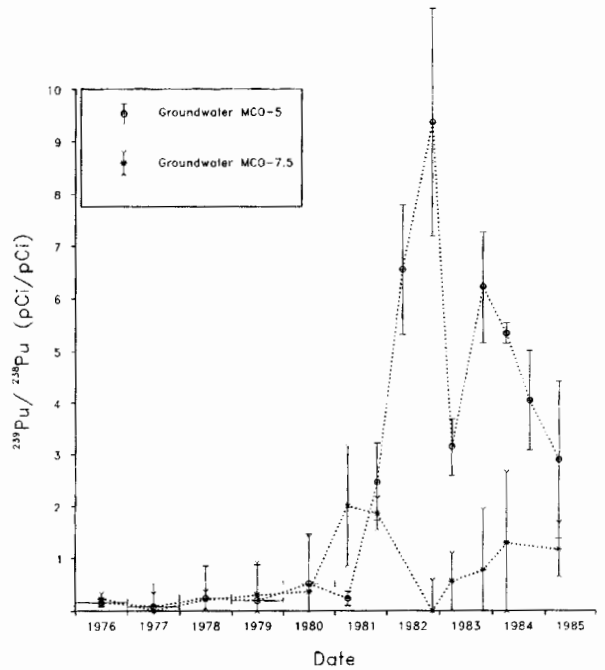

FIGURE 3. Plutonium isotopic ratios in groundwater at MCO-7.5 and MCO-5. The arrival of isotopically heavy plutonium in the downgradient well before its arrival in the upgradient well shows that this plutonium could not have been transported through groundwater (data from refs 3, 15, 16, and 29−35).

hypotheses for the movement of plutonium into monitoring wells (Supporting Information). Data on ^{241}Am in groundwater and alluvium, which would be directly comparable to earlier work, are too incomplete to be useful in testing possible mechanisms, and these data are not presented here.

Effluent has been sampled routinely at the outfall to the canyon and includes average annual activity of ^{238}Pu, ^{239}Pu, and tritium released to the canyon. The values utilized in this study are averages for the year. Quantitative error estimates are not reported for this information.

Surface water and perched groundwater have been monitored annually or semi-annually at a surface water gaging station (GS-1, Figure 1) and at six monitoring sites (MCO-3, MCO-4, MCO-5, MCO-6, MCO-7, and MCO-7.5). These monitoring wells have been used in all previous studies of shallow groundwater in Mortandad Canyon. Wells constructed before 1974 (MCO-3, MCO-4, MCO-5, and MCO-7) were sealed with cuttings, and wells constructed after 1974 (MCO-6 and MCO-7.5) were gravel packed (36; Table 1). None of the monitoring wells have bentonite or grout seals.

Error values reported for groundwater analyses are twice the standard deviation of the distribution of observed values except for results from single analyses where the error is twice the analytical uncertainty. Error values exceed mean values for some determinations, but these determinations were retained in the data set as evidence that the concentrations of the analyte was less than the detection limit (taken as the mean value plus the error value). The exact collection dates for samples collected between 1976 and 1980 are uncertain, and this uncertainty is incorporated in the discussion.

Samples of surface alluvium have been collected annually or semi-annually from three sites in the portion of the canyon containing shallow groundwater (at GS-1 near MCO-3, at MCO-5, and at MCO-7; Figure 1). Error values for analyses represent twice the standard deviation of the distribution of observed values except for single analyses where error values are twice the analytical uncertainty. Error values were smaller than the mean value for all but one of the determinations, and the determination where the error value exceeded the

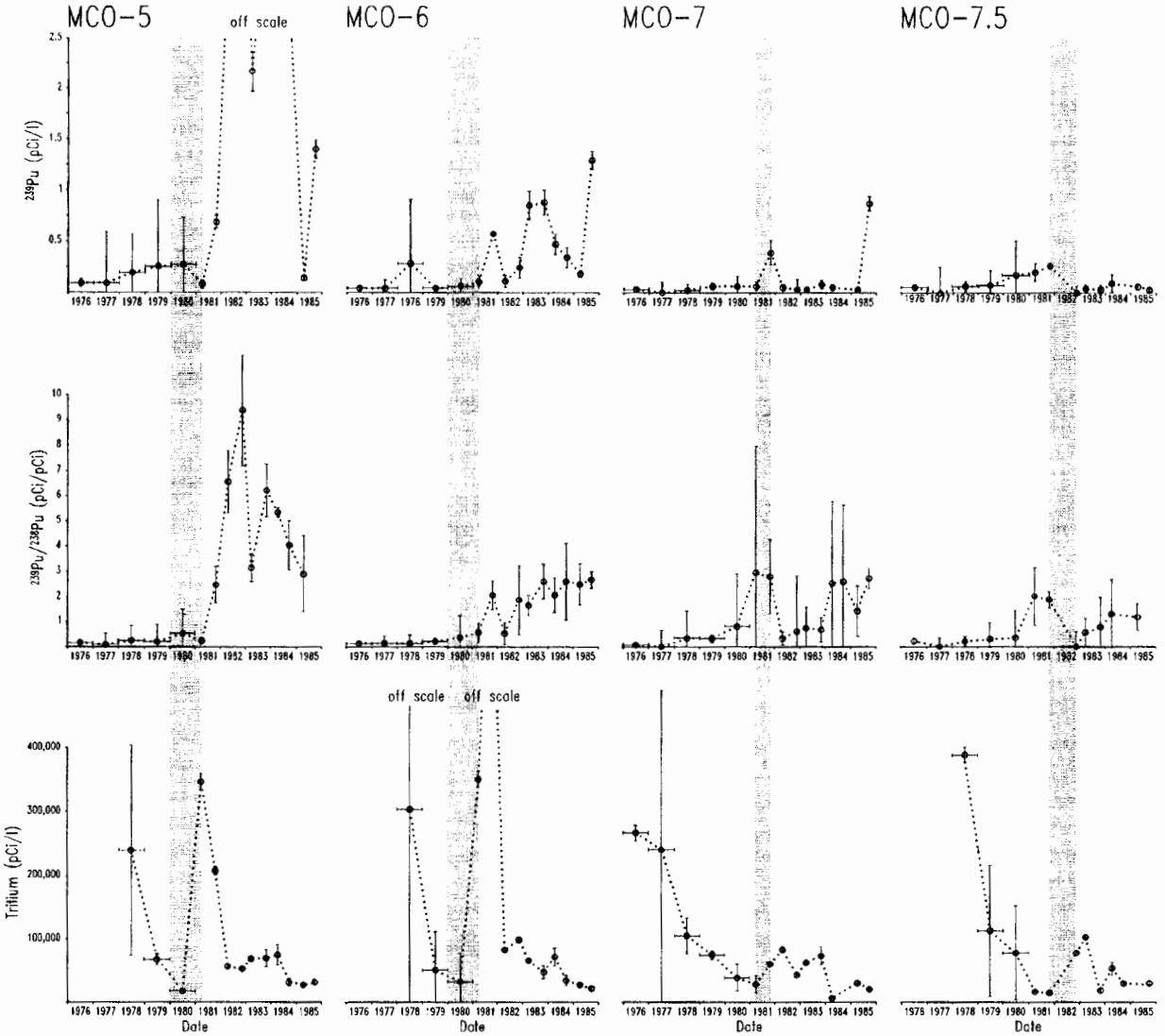

FIGURE 4. ^{239}Pu, ^{239}Pu/^{238}Pu ratios, and tritium in monitoring wells of Mortandad Canyon. Shaded intervals show the arrival of high tritium groundwater in the monitoring wells. Groundwater rich in ^{239}Pu and with a high ^{239}Pu/^{238}Pu ratio arrived after high tritium groundwater in wells MCO-5 and MCO-6, at the same time as high tritium groundwater in well MCO-7, but long before high tritium groundwater in well MCO-7.5 (data from refs 3, 15, 16, and 29–35).

mean value was treated as described above for water samples. The exact collection date of alluvium samples is uncertain, and this uncertainty is incorporated in the discussion.

Errors were propagated into derived quantities, such as ratios, using standard methods (*37*). Propagated error values for derived quantities represent 95% confidence intervals.

Discussion

Plutonium in effluent discharged to Mortandad Canyon before 1968 was dominated by ^{239}Pu produced in weapons and reactor research, and during this period the ^{239}Pu/^{238}Pu ratio in alluvium and shallow groundwater was considerably greater than 1 (*38*). Between 1968 and 1978, ^{238}Pu in effluent increased sharply as research on space reactors and biomedical applications increased, and during this period ^{239}Pu/^{238}Pu ratios in surface alluvium and shallow perched groundwater fell to values considerably less than 1 (*38*; Supporting Information). ^{239}Pu/^{238}Pu in effluent began to shift upward once again in 1978, but relatively little plutonium entered the canyon until 1980 when the amount of plutonium discharged to the canyon and the ^{239}Pu/^{238}Pu ratio both increased sharply (Figure 2). Tritium concentrations in effluent also attained a short-term high at about the same time.

Plutonium transported through shallow groundwater by colloidal transport should behave differently from plutonium, which is eroded, carried in surface runoff, and deposited in wells from the surface. The large amount of isotopically distinct plutonium discharged during the early 1980s is ideal for testing hypotheses about plutonium transport in Mortandad Canyon. The four interrelated lines of inquiry best suited for this purpose are as follows:

(1) Determination of the rate of transport of high ^{239}Pu–high ^{239}Pu/^{238}Pu water down canyon and comparison of this rate to the expected rate of groundwater flow determined in earlier studies.

(2) Comparison of the arrival patterns of high tritium and high ^{239}Pu–high ^{239}Pu/^{238}Pu groundwater in monitoring wells.

(3) Determination of arrival times and isotopic ratios of high ^{239}Pu/^{238}Pu plutonium in co-collected samples of groundwater and surface alluvium.

(4) Examination of plutonium/tritium ratios in groundwater through time.

Transport Rates Determined from the First Appearance of ^{239}Pu-Rich Plutonium in Monitoring Wells. Earlier studies suggested that the rate of plutonium movement may be too rapid to be explained by groundwater transport (*4*). Colloidal

transport should move plutonium at approximately the rate of groundwater flow, but movement of plutonium in surface water potentially could be much more rapid.

High $^{239}Pu/^{238}Pu$ plutonium was detected in well MCO-7.5 on April 2, 1981 (2.0 ± 1.1 pCi/pCi; Figure 3; Supporting Information), while $^{239}Pu/^{238}Pu$ ratios in upstream monitoring wells remained low at this time (0.23 ± 0.13 pCi/pCi in MCO-5, for example). The detection in MCO-7.5 is not a one time event because the ratio remained high in the subsequent sampling event (1.86 ± 0.30 pCi/pCi). This detection of plutonium in downgradient wells before upgradient wells is difficult to reconcile with the movement of plutonium through groundwater.

If the anomalous appearance of contamination in the downgradient well is ignored, the rate of contaminant movement in the aquifer can be determined. High $^{239}Pu/^{238}Pu$ plutonium was first detected in wells MCO-5 and 6 on October 26, 1981 (2.46 ± 0.74 and 2.04 ± 0.58 pCi/pCi, respectively; Figure 4). Isotopic ratios for wells MCO-4 and 7 for April 2, 1981, have high uncertainties (3.67 ± 10.56 and 2.95 ± 5.02, respectively), and it is not possible to determine directly when high $^{239}Pu/^{238}Pu$ plutonium entered the wells. ^{239}Pu concentrations, however, increased in concert with isotopic ratio in the other wells, and ^{239}Pu did not increase in wells MCO-4 and MCO-7 until October 26, 1981, when it rose from 0.11 ± 0.12 to 3.4 ± 0.28 pCi/L and from 0.056 ± 0.036 to 0.39 ± 0.12 pCi/L, respectively (Figure 4; Supporting Information).

$^{239}Pu/^{238}Pu$ also was high in well MCO-7.5 on October 26, 1981 (1.86 ± 0.30), and if the anomalous first appearance of high $^{239}Pu/^{238}Pu$ groundwater in this well is ignored, high ^{239}Pu, high $^{239}Pu/^{238}Pu$ groundwater entered five monitoring wells between April 2 and October 26, 1981. This would require movement of contaminated water over the 428 m separating wells MCO-4 and -5, the 425 m separating wells MCO-5 and -6, and the 730 m separating wells MCO-6 and -7.5 within 207 days (an average transport rate of at least 7.6 m/day). Groundwater flows approximately 18 m/day between wells MCO-4 and -5, 6 m/day between wells MCO-5 and -6, and 2 m/day east of monitoring well MCO-6 and would require approximately 460 days to cover the distance between the wells (an average transport rate of 3.4 m/day). Transport of plutonium, therefore, was approximately twice as rapid as groundwater flow.

Plutonium contamination does not always move rapidly between wells, however. In the initial sampling event of 1982, for example, the activity of ^{239}Pu in wells MCO-3, MCO-4, and MCO-5 increased by factors of 100, 4, and 5, respectively, above levels observed in the final sampling event of 1981 (Figure 4). The concentration of ^{239}Pu in these wells remained high through 1982, but this spike in contamination never appeared in downgradient wells MCO-6, MCO-7, or MCO-7.5. All 1982 ^{239}Pu results for the downgradient wells, in fact, fell significantly from levels observed at the end of 1981. This clearly shows that the transport of plutonium is episodic and not uniform.

Arrival of High Tritium and High Plutonium in Monitoring Wells. Tritium in groundwater moves without significant retardation on aquifer materials and moves at the rate of groundwater flow. A tritium spike moved through the groundwater of Mortandad Canyon during the early 1980s, and the rates of tritium and plutonium transport during this period can be compared directly.

Tritium levels in the upgradient monitoring wells MCO-4, -5, and -6 increased sharply in the April 2, 1981, sampling event, before isotopically heavy plutonium appeared in the wells (Figure 4). Tritium increased in well MCO-7 in the October 26, 1981, sampling event at the same time $^{239}Pu/^{238}Pu$ increased for the first time. The high tritium groundwater, however, was not detected in well MCO-7.5 until November 15, 1982, long after the initial increase in $^{239}Pu/^{238}Pu$ ratios in this well (Figure 4). This confirms the observation that initial plutonium enters the downgradient wells more rapidly than can be explained by groundwater transport.

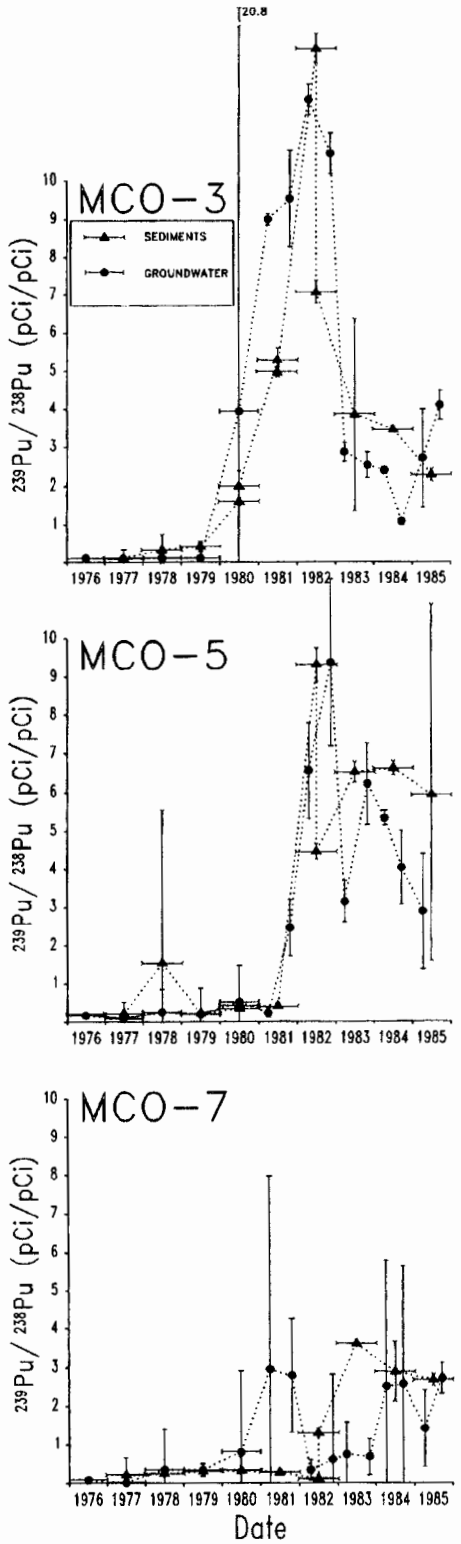

FIGURE 5. Comparison of $^{239}Pu/^{238}Pu$ ratios in co-collected alluvium and groundwater samples. Isotopic ratios in groundwater and surface alluvium show linkage between plutonium in surface alluvium and groundwater plutonium for wells MCO-3 and -5 (data from refs 3, 15, 16, and 29–35).

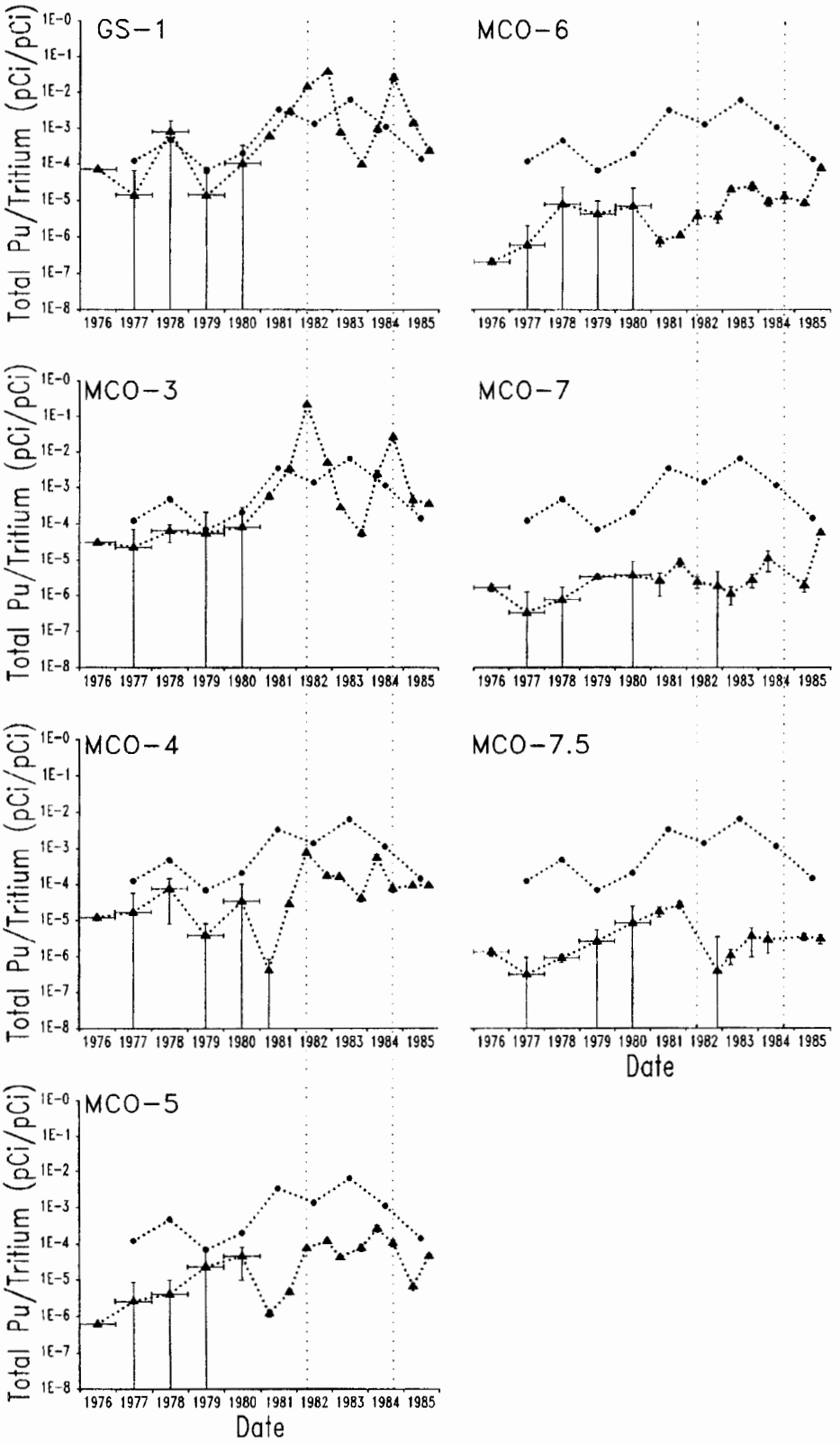

FIGURE 6. Plutonium to tritium ratios in monitoring wells. The dashed lines show the timing of peak plutonium to tritium ratios in well MCO-3. Ratios greater than those of the effluent show that plutonium was concentrated before it was injected into the well. Periods of plutonium injection into the upgradient monitoring wells do not effect the downgradient wells (data from refs 3, 15, 16, and 29–35).

Moreover, the appearance of tritium in monitoring well MCO-7.5 was not associated with either an increase in ^{239}Pu or a further increase in the $^{239}Pu/^{238}Pu$ ratio. In fact, ^{239}Pu in monitoring well MCO-7.5 fell below detection levels during the November 15, 1982, event, and ^{239}Pu values remained substantially below 1981 levels throughout 1983 (Figure 4).

This shows that the plutonium detected in groundwater in the upgradient wells was not transported down canyon with the groundwater.

Plutonium Isotope Ratios in Surface Alluvium and Groundwater. Data on co-collected surface alluvium and groundwater are available for three sites in Mortandad Canyon MCO-3/GS-1, MCO-5, and MCO-7 (Figure 1). Colloidal transport would not predict any direct link between plutonium in surface alluvium and shallow groundwater, but transport through surface runoff and subsequent contamination of wells would require surface alluvium to receive plutonium before groundwater.

The relationship between $^{239}Pu/^{238}Pu$ ratios in surface alluvium and groundwater for well MCO-7 are obscured by analytical uncertainties, but there is no clear link between the two sets of values (Figure 5). The correlation between isotopic ratios in alluvium and groundwater, however, is striking for the other two wells (Figure 5). At MCO-3 and MCO-5, high $^{239}Pu/^{238}Pu$ plutonium arrived simultaneously in the surface alluvium and in groundwater within the resolution of the available data. Moreover, the correlation coefficients for isotopic ratios in the alluvium and groundwater are 0.92 and 0.92, respectively, for average annual values between 1977 and 1985, and the highest $^{239}Pu/^{238}Pu$ ratios attained in surface alluvium and groundwater are virtually identical (13.47 ± 0.36 and 12.14 ± 0.40 pCi/pCi, respectively, for MCO-3 and 9.31 ± 0.40 and 9.37 ± 2.18 pCi/pCi, respectively, for MCO-5). The correlation of the maximum $^{239}Pu/^{238}Pu$ values in surface alluvium and sediments show that plutonium in both reservoirs was diluted by the same factor as it moved down canyon and suggests that the two reservoirs of plutonium are not separated.

Comparison of Plutonium and Tritium in Groundwater. TA-50 is the only source of radioactive contamination to Mortandad Canyon, and the departure of plutonium to tritium ratios from effluent values provides a convenient measure of the relative enrichment and relative depletion of plutonium in groundwater. Plutonium is depleted relative to effluent in all sampling events at all wells with the exception of well MCO-3 (Figure 6). The plutonium to tritium ratios in well MCO-3 periodically exceeded effluent values. Enrichment reached 154-fold and 25-fold during the April 21, 1982, and September 19, 1984, sampling events, respectively; values that are too large to be explained by short-term temporal variation in effluent composition.

Plutonium must have been concentrated relative to tritium before moving into this monitoring well. Plutonium entering the canyon is concentrated initially on alluvium near the effluent outfall, and mobilization of plutonium from these sediments probably is responsible for the observed injection events. Erosion of sediment and transport of the sediments to the monitoring well is the most likely mechanism for mobilization, and while other possible mechanisms for mobilization cannot be entirely discounted, the correlation of plutonium to tritium ratios in groundwater at MCO-3 and in surface water at GS-1 support this interpretation.

The increase in the plutonium to tritium ratio seen in well MCO-3 and surface water between April 1981 and April 1982 affected wells MCO-4 and -5 to a lesser degree, and wells MCO-6, -7, and -7.5 remained unaffected. In general, plutonium/tritium ratios fluctuate less in wells further down the canyon. The highest and lowest plutonium to tritium ratios recorded between 1976 and 1986 differ by a factor of 9500 for well MCO-3, for example, but only by a factor of 85 for well MCO-7.5. Since tritium moves into and out of the monitoring wells with water, this difference shows that movement of plutonium into and out of the shallow upgradient wells MCO-3, -4, and -5 is relatively easy as compared to deeper downgradient wells. This relationship would not be expected if plutonium moved into all wells on colloids transported in groundwater.

Significance. The detection of plutonium with high $^{239}Pu/^{238}Pu$ ratios in a downgradient monitoring well before its detection in wells closer to the outfall shows that contamination could not have entered the downgradient well through the groundwater. The initial high $^{239}Pu/^{238}Pu$–high ^{239}Pu contaminant front, moreover, moved between wells MCO-4 and MCO-7.5 at least twice as fast as the expected groundwater flow velocity. The front arrived after a distinctive tritium spike in upgradient wells but along with the spike in well MCO-7 and long before the spike in well MCO-7.5. Such transport also shows that plutonium could not have moved in the subsurface and argues that plutonium must have moved across the surface before entering the monitoring wells.

Plutonium isotopic ratios in surface alluvium and groundwater at wells MCO-3/GS-1 and MCO-5 are linked and suggest that plutonium moved quickly from the surface into the monitoring wells of the upper canyon. This is bolstered by plutonium to tritium ratios, which show that plutonium was concentrated relative to tritium and periodically injected into monitoring wells of the upper canyon and that plutonium enters and leaves the upgradient monitoring wells readily.

Leakage of contaminant-bearing surface waters into the subsurface has been widely documented in wells that lack impermeable seals (*39*), and old, poorly constructed monitoring wells may contribute to the rapid movement of plutonium from the surface into the shallow groundwater. The available data, however, cannot rule out the possibility that plutonium contamination may move downward from the surface into the shallow perched groundwater through physical mixing in the stream bed and percolation over short distances as proposed by Baltz et al. (*40*).

Surface water flow is less frequent and less voluminous in the lower canyon, and the wells are screened deeper and are of more recent construction (Table 1). These factors appear to be at least partially responsible for the relatively infrequent movement of plutonium contamination into the wells in the lower canyon. The better isolation of these wells from the surface also may be responsible for preferentially excluding large colloids from the wells producing the observed variation in apparent americium distribution coefficients interpreted previously as evidence of colloidal transport.

Transport of plutonium over long distances in Mortandad Canyon is cited frequently as an example of the potential significance of colloidal transport in contaminant migration (*7, 9, 41–51*). Colloidal transport of plutonium over short distances (meters or tens of meters) in the canyon cannot be discounted by the current discussion, but such transport clearly has not moved plutonium over large distances as claimed. The threat of plutonium contamination of groundwater through this mechanism, therefore, may be overstated significantly, and a rigorous assessment of potential plutonium migration through colloidal transport at other sites could permit resources to be shifted away from monitoring plutonium in groundwater and toward actual threats.

Acknowledgments

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Supporting Information Available

Three tables giving data on the radionuclides in effluent entering Mortandad Canyon from TA-50, the concentrations

of radionuclides in surface water and groundwater, and the radionuclides in surface alluvium of Mortandad Canyon (6 pp) will appear following these pages in the microfilm edition of this volume of the journal. Photocopies of the Supporting Information from this paper or microfiche (105 × 148 mm, 24× reduction, negatives) may be obtained from Microforms Office, American Chemical Society, 1155 16th St. NW, Washington, DC 20036. Full bibliographic citation (journal, title of article, names of authors, inclusive pagination, volume number, and issue number) and prepayment, check or money order for $16.50 for photocopy ($18.50 foreign) or $12.00 for microfiche ($13.00 foreign), are required. Canadian residents should add 7% GST. Supporting Information is also available via the World Wide Web at URL http://www.chemcenter.org. Users should select Electronic Publications and then Environmental Science and Technology under Electronic Editions. Detailed instructions for using this service, along with a description of the file formats, are available at this site. To download the Supporting Information, enter the journal subscription number from your mailing label. For additional information on electronic access, send electronic mail to si-help@acs.org or phone (202)872-6333.

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