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Influence of sediment storage on downstream delivery of contaminated sediment

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[1] Sediment storage in alluvial valleys can strongly modulate the downstream migration of sediment and associated contaminants through landscapes. Traditional methods for routing contaminated sediment through valleys focus on in-channel sediment transport but ignore the influence of sediment exchanges with temporary sediment storage reservoirs outside the channel, such as floodplains. In theory, probabilistic analysis of particle trajectories through valleys offers a useful strategy for quantifying the influence of sediment storage on the downstream movement of contaminated sediment. This paper describes ā field application and test of this theory, using ^{137}Cs as a sediment tracer over 45 years (1952–1997), downstream of a historical effluent outfall at the Los Alamos National Laboratory (LANL), New Mexico. The theory is parameterized using a sediment budget based on field data and an estimate of the ^{137}Cs release history at the upstream boundary. The uncalibrated model reasonably replicates the approximate magnitude and spatial distribution of channel- and floodplain-stored ^{137}Cs measured in an independent field study. Model runs quantify the role of sediment storage in the long-term migration of a pulse of contaminated sediment, quantify the downstream impact of upstream mitigation, and mathematically decompose the future ^{137}Cs flux near the LANL property boundary to evaluate the relative contributions of various upstream contaminant sources. The fate of many sediment-bound contaminants is determined by the relative timescales of contaminant degradation and particle residence time in different types of sedimentary environments. The theory provides a viable approach for quantifying the long-term movement of contaminated sediment through valleys.

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1. Introduction

[2] Chemicals in the environment commonly bind to soils and sediments, which are ultimately carried by rivers. The movement of contaminated particles through fluvial systems largely determines the fate of sediment-bound pollutants. Empirical studies over the past several decades have demonstrated that much of the sediment delivered to rivers enters short- or long-term storage in deposits such as the channel bed, bars, and floodplains. Floodplain storage of sediment in particular is recognized as an important component of the sediment budget in many fluvial systems [e.g., *Meade*, 1982; *Kesel et al.*, 1992; *Dunne et al.*, 1998]. A number of studies have demonstrated that floodplains can be dominant sources or sinks of sediment-bound contaminants in rivers [e.g., *Graf et al.*, 1991; *Marron*, 1992; *Miller et al.*, 1999; *Coulthard and Macklin*, 2003]. Temporary storage of sediment in alluvial sediment storage reservoirs must influence the downstream delivery of particle-bound pollution, though this effect has not been quantified.

[3] Despite observations that sediment exchange with floodplains is significant in many alluvial valleys, traditional approaches to routing sediment through valleys have focused on in-channel processes such as sediment transport, erosion, and deposition. Exchanges with the floodplain are usually ignored or treated qualitatively [*Vanoni*, 1975]. As a result, the capability for quantitatively predicting the fate of sediment and associated constituents residing in floodplains is limited, even though floodplains contain most of the sediment and contaminants in many valley floors. Probability theory has been proposed as a strategy for analyzing the role of temporary storage in the downstream routing of sediment in river valleys [*Dietrich et al.*, 1982; *Kelsey et al.*, 1987]. This approach was formalized in an earlier paper [*Malmon et al.*, 2003], and can be parameterized using a sediment budget of the valley floor. This technique offers considerable potential as a means of quantifying the role of sediment storage in the long-term movement of sediment and associated contamination through alluvial valleys.

[4] The theoretical framework for stochastic modeling of particle trajectories has been established [*Malmon et al.*,

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2003], but it has yet to be rigorously tested in a field setting. This paper provides a field test of the method, using the particle-bound radionuclide ^{137}Cs as a sediment tracer in an alluvial valley at Los Alamos National Laboratory (LANL), New Mexico. Sediment trajectories are modeled by considering the frequency and duration of particle storage in channels and floodplains. The probabilistic model is parameterized using a sediment budget based on field measurements [*Malmon*, 2002; *Malmon et al.*, 2004]. The model simulates the redistribution and radioactive decay of particle-bound ^{137}Cs discharged into upper Los Alamos (ULA) Canyon over several decades. We test the model by comparing the modeled distribution of ^{137}Cs in 1997 with an independent estimate determined for that year based on geomorphic mapping and sampling (based on field data from *Reneau et al.* [1998] and *LANL* [2004]). We conduct numerical simulations to predict the fate of ^{137}Cs currently residing in the valley, and quantify the impact of remediation on the downstream delivery of sediment-bound contamination from the watershed. The paper concludes with a discussion of the extent to which this approach is transferable to other rivers.

2. Field Area

[5] The ULA Canyon watershed (28 km^2 above its confluence with Pueblo Canyon) drains a mainly volcanic landscape that contains portions of the Jemez Mountains and the Pajarito Plateau [*Smith et al.*, 1970]. The watershed (Figure 1) contains contamination resulting from point source discharges of low-level radioactive liquid waste. One of the most important of the contaminants from the perspective of potential human health risk is ^{137}Cs [*Reneau et al.*, 1998], which binds to sediment and decays radioactively with a half-life of 30.2 years. The main source of ^{137}Cs was a wastewater treatment plant at LANL Technical Area (TA) 21 that discharged into DP Canyon, a major tributary to ULA Canyon at the head of the study reach (Figure 1). Effluent releases at that site occurred from 1952 through 1986 [*Katzman et al.*, 1999]. Cesium-contaminated sediment has since been dispersed throughout DP and Los Alamos Canyons by fluvial processes.

[6] For the purpose of the model, the Los Alamos Canyon valley below DP Canyon is divided into four reaches, varying in length between 0.7 and 1.6 km (Figure 1). The average channel gradient in these reaches is 0.02, and there are no major changes in gradient. Several factors influence valley geometry in the study area, including a base level set by basalt outcrops near the confluence with Pueblo Canyon and a large bouldery deposit (dated at circa 1300–1650 A.D. by *Reneau and McDonald* [1996]) that occupies much of the space in the valley floor. The active portion of the valley floor is inset into the bouldery unit and is usually between 5 and 20 m wide, containing an ephemeral channel averaging 1.9 m wide (Figure 2).

[7] Sediment is stored beneath the channel bed and within floodplain deposits along the channel margins (Figure 2b). While there are significant local variations in the texture of the channel bed, in general it is composed of particles of medium sand through gravel (particles with diameter 0.25 mm or larger). The floodplain deposits are dominated by finer-grained sediment, generally consisting of medium sand and finer particles (particles smaller than 0.5 mm) [*Reneau et al.*, 1998]. The particle size distributions of the channel and floodplain correspond well with those of the bed load and suspended load, respectively [*Malmon et al.*, 2004]. Because these two components of the sediment load are generally supplied from different sources, transported by different mechanisms, and stored in different types of deposits, the model treats the two classes of sediment as separate populations of particles with different trajectories through the valley floor. The distinction is also important because ^{137}Cs adsorbs preferentially onto fine sediment.

3. Model Development and Parameterization

3.1. Conceptual Model

[8] The trajectory of a particle through the fluvial system consists of a series of short-duration hops separated by periods of temporary storage in sediment storage reservoirs such as channel bars and floodplains. Each of these hops has an annual probability called the transition probability. For reasonably well mixed sediment reservoirs, *Malmon et al.* [2003] show how the transition probabilities can be computed from (1) the masses of the sediment reservoirs, and (2) the annual rates of downstream sediment transport and exchange among the temporary storage reservoirs. Collectively, these two sets of quantities are known as the sediment budget of a valley floor [*Dietrich et al.*, 1982] and can be estimated using a variety of empirical and theoretical methods [e.g., *Reid and Dunne*, 1996]. For the sake of brevity, the equations for computing and analyzing the transition probabilities are not included in this paper. For a more thorough explanation of the theory, we refer the reader to the original paper [*Malmon et al.*, 2003].

[9] The model framework developed for ULA Canyon is depicted in Figure 3, which shows all the possible particle exchanges that can occur in a given year. The sediment reservoirs are referred to as transient states, and sediment transported past the downstream boundary is said to have entered the absorbing state. The model shown in Figure 3 does not allow particles to move between the channel bed and the floodplain, reflecting the interpretation by *Malmon et al.* [2004] that the sediment load is partitioned into two distinct fractions that are transported by different mechanisms and stored in distinct locations. A previous, simpler version of the model [*Malmon et al.*, 2002] permitted sediment exchanges between the channel bed and the floodplain by assuming that all particles were suspendible and, once mobilized, could be deposited in either the channel or floodplain. The current version of the model only allows coarse sediment to deposit in the channel and fine sediment in the floodplain. This conceptual model is more realistic because it differentiates between the two fractions of the sediment load for the reasons noted above.

[10] The model depicted in Figure 3 ignores a component of the fine sediment in the channel bed and a component of coarse sediment in the floodplain. While both of these have been noted in the field they were judged to be second-order influences on long-term sediment migration through the valley floor. Fine particles (<0.25 mm) account for less than 15% of the channel bed sediment [*Reneau et al.*, 1998]; these are probably deposited during the waning stages of flow and remobilized quickly during subsequent events, and

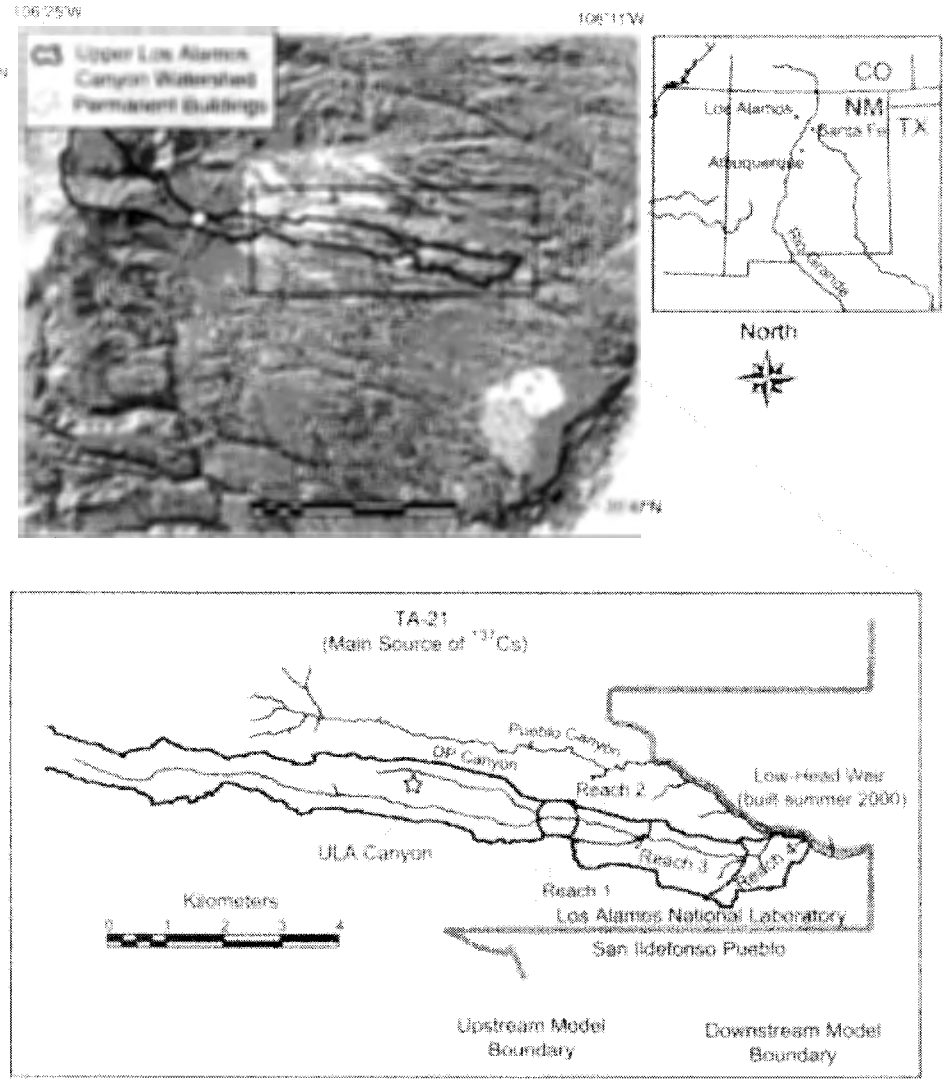

Figure 1. Location map of upper Los Alamos (ULA) Canyon and the Pajarito Plateau, in north central New Mexico. Shaded relief map shows watershed boundary, the extent of urbanization on the plateau, and the location of Los Alamos Reservoir. Inset map depicts the extent of the four modeling reaches, the property boundary between Los Alamos National Laboratory and the San Ildefonso Pueblo, location of the primary point source of ^{137}Cs in the DP Canyon watershed, and the location of an engineered barrier ("low-head weir") constructed in summer 2000 to deposit ^{137}Cs-contaminated sediment upstream of the LANL property boundary.

thus probably have little impact on the long-term predictions of the model. Coarse particles (>0.5 mm) account for about 25% of the floodplain sediment. This portion comprises primarily low-density volcanic sand and gravel deposited overbank during large floods. Additionally, some of the coarse sediment in the floodplain may be postdepositional material mixed vertically upward by burrowing animals. The model does not specifically account for either of these mechanisms. While the quantity of coarse sediment in the floodplain is not negligible, these particles, once mobilized, are subsequently transported and stored in the same manner as other coarse sediment. Thus these mechanisms probably reduce the mobilization probabilities for coarse particles but do not affect the deposition probabilities. Ignoring the mechanisms that deposit coarse sediment in the floodplain should cause the model to predict more rapid downstream sediment migration compared with a model that included them, however the quantitative impact of this omission is not known.

[11] We assume that sediment storage in the valley floor is approximately in a steady state condition over timescales relevant to the analysis, namely several decades. This approximation greatly simplifies the analysis and reduces the amount of data necessary for parameterization. The

A

B

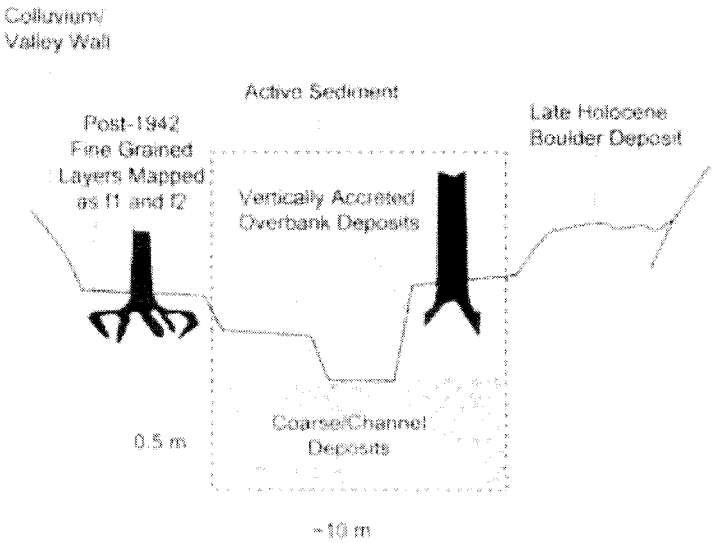

Figure 2. Configuration of the valley floor. (a) Photograph of reach LA 2 East in LLA Canyon, containing a sand and gravel intermittent channel with narrow floodplain underlain by finer sediment. The photograph was taken summer 1996 during an independent field study measuring the distribution of ^{137}Cs in the valley floor [*Reneau et al.*, 1998]. The person is conducting a gross gamma radiation walkover survey. (b) Two distinct facies of deposits found in the active sediment of the valley floor: (1) coarse sediment underlying the channel bed and representing the bed material load during floods and (2) vertically accreted fine-sediment deposits overlying the coarse deposits and forming narrow floodplains. The recent fine-grained layers mapped as f1 and f2 (outside the box labeled "Active Sediment") lie outside the model state space; sediment exchange with these higher floodplain units is not modeled explicitly. Modified from *Malmon et al.* [2004].

assumption is based on (1) LANL ground photographs that indicate little change in channel morphology since the 1940s, (2) observations that abandoned channel bed deposits beneath the floodplains typically lie at approximately the same elevation as the present channel bed, indicating generally little change in channel bed elevation, and (3) thicknesses of fine sediment on the floodplain are symmetrically distributed, as would be expected for a floodplain in steady state

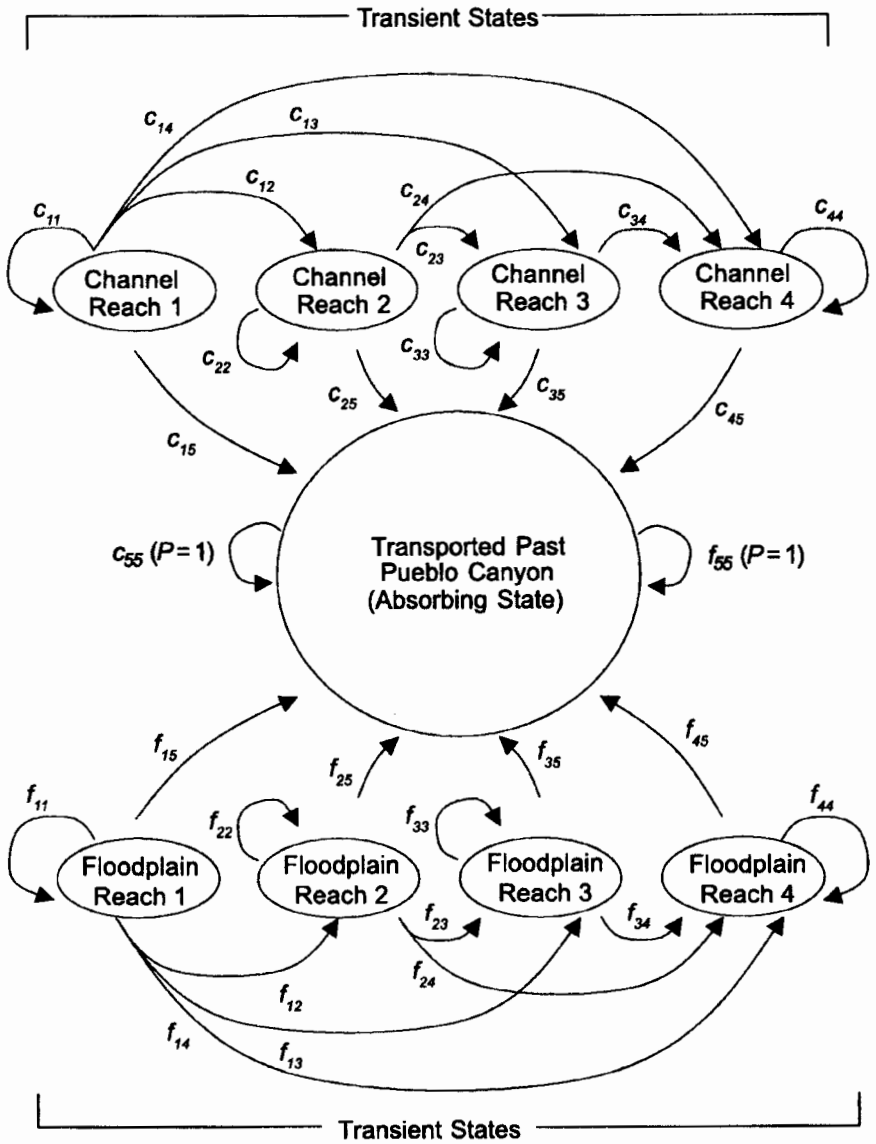

Figure 3. Model of particle trajectories through four reaches of ULA Canyon. This model recognizes two distinct classes of sediment: (1) coarse sediment, which can be exchanged with the channel bed (via transitions c_{ij}), and (2) fine sediment, which interacts with the floodplain (transitions f_{ij}). The model is specified by two separate transition matrices, **C** and **F** (Tables 2a and 2b). The transition probabilities p_{ij} can be computed from the values in the sediment budget (see text for explanation).

[*Malmon*, 2002, p. 98]. While the steady state assumption may be valid locally and in many other field areas, it is an important condition that limits the ability of the model to be used without modification in many other settings, such as aggrading rivers. *Malmon et al.* [2003, pp. 540–541] further discuss the possibility of adapting the approach to transient sediment storage conditions.

3.2. Sediment Budget

[12] The sediment budget of the valley floor, computed by *Malmon* [2002] and *Malmon et al.* [2004] using field data and simple process models, is summarized in Table 1. The sediment budget considers sediment storage in two reservoirs (the channel and the floodplain), along four contiguous reaches of valley floor. Two distinct transport mechanisms were quantified for the coarse- and fine-sediment fractions using relationships based on field data collected from flash floods during the summer monsoon season [*Malmon et al.*, 2004]. These relationships were integrated over a probability distribution of hydrographs (derived from rainfall-runoff modeling calibrated to basin hyetographs and hydrographs [*Malmon*, 2002]) to determine the long-term average fluxes of coarse and fine sediment. The sediment budget also contains rates of

Table 1. Estimated Sediment Budget of Upper Los Alamos Canyon

	Reach 1	Reach 2	Reach 3	Reach 4
Mass of sediment in the channel bed,[a] t	3100	9400	6200	5000
Floodplain mass,[b] t	2000	6500	3600	2700
Rate of channel bed erosion/deposition,[c] t/yr	2500	5600	6100	5000
Rate of overbank sedimentation/bank erosion,[d] t/yr	40	130	70	50
Bed load,[e] t/yr	1100	1100	1100	1100
Suspended load,[e] t/yr	1100	1100	1100	1100

[a]Assumes bulk density for channel sediment is 1.23 g/cm^3, based on data from *Reneau et al.* [1998]. Active channel is assumed to be 0.5 m thick, corresponding to the approximate maximum depth of scour during the largest observed flow events, measured using scour chains from 1998 to 2000. Active coarse sediment in channel bed is assumed to extend beneath the floodplain to an average depth of 0.5 m (see Figure 2b).

[b]Assumes bulk density for floodplain sediment is 1.04 g/cm^3, based on data from *Reneau et al.* [1998]. Active floodplain area is the area mapped as c2 and c3 by *Reneau et al.* [1998] and *LANL* [2004]. Floodplain thickness is assumed to be 0.5 m, the area-weighted average mean thickness of fine sediment on units mapped as c2 and c3 in sampling reaches LA-2 East and LA-3 [*Reneau et al.*, 1998].

[c]Refers to the modeled annual rate of channel bed erosion and deposition (they are equal to one another because channel is neither aggrading or degrading and therefore assumed to be in steady state). This value was computed by *Malmon* [2002] using an empirical event scour model based on scour chain data and integrated over the probability distribution of modeled hydrographs.

[d]Based on a vertical sedimentation rate of 1 cm/yr, derived from dendrochronological and stratigraphic data [*Reneau et al.*, 1998; *LANL*, 2004], and an overbank sedimentation model applied to the probability distribution of flow [*Malmon*, 2002]. Mass rate of bank erosion is assumed to be equal to the floodplain sedimentation rate based on the steady state assumption (see text for explanation).

[e]Annual suspended and bed load fluxes from rating curves based on field measurements during thunderstorm-generated flash floods and integrated over the probability distribution of flow events [*Malmon et al.*, 2004].

exchange between the flow and the floodplain (by overbank sedimentation and bank erosion) and between the flow and the channel bed (by vertical scour and fill of the bed). The notes in Table 1 briefly describe the methodology used to obtain the estimated values.

[13] The transition probabilities were computed from the values in Table 1 using previously published equations [*Malmon et al.*, 2003]. The transition probabilities were arranged in two matrices, **C** and **F** (Tables 2a and 2b), corresponding to coarse (channel) sediment and fine (floodplain) sediment. The calculations in the rest of this paper are based on simple manipulations of the matrices in Tables 2a and 2b [*Malmon et al.*, 2003].

3.3. Cesium-137 Input History

[14] In order to test the model using ^{137}Cs as a tracer, it is necessary to estimate the amount of tracer input into the system over time. Among the facilities in the ULA watershed was an industrial wastewater treatment plant at TA-21 that discharged low-level radioactive effluent into DP Canyon (Figure 1) beginning in 1952 and ending in 1986. On the basis of Department of Energy records, *Stoker et al.* [1981, p. 29] reported a total release of 18 millicuries (mCi) of ^{137}Cs at TA-21. However, more recent investigations estimated that ^{137}Cs inventories in DP and ULA Canyons were approximately 109 and 275 mCi, respectively, in 1997 [*Reneau et al.*, 1998; *Katzman et al.*, 1999; *LANL*, 2004]. Therefore the release records are incomplete and unreliable for reconstructing a release history, other than to define the approximate timing of release of various radionuclides. Furthermore, because the model requires the ^{137}Cs input at the mouth of DP Canyon, not at the contaminant source at TA-21, we used sedimentologic evidence in the lower portion of DP Canyon rather than estimates of contaminant release at the outfall to define the ^{137}Cs input history. This approach makes the following assumptions: (1) that decay-corrected ^{137}Cs concentrations in samples of datable deposits of coarse and fine sediment near the mouth of DP Canyon are representative of concentrations on sediment discharged from DP Canyon the time the sediments were deposited; and (2) that postdepositional disturbances that would affect ^{137}Cs concentrations (other than radioactive decay) have been minimal. *Malmon* [2002, Appendix C] detailed these analyses and tabulated the original data.

[15] The reconstructed history of ^{137}Cs concentration on sediment leaving DP Canyon is plotted in Figure 4. Cesium discharges began in 1952, and peaked sometime in the late 1950s [*Reneau et al.*, 1998; *Katzman et al.*, 1999]. Cesium concentrations on sediment in transport (both suspended load and bed load) and in recent deposits (both fine- and coarse-grained) generally vary between 1 and 10 pCi/g, and appear to be decreasing [*Malmon et al.*, 2002].

[16] Contaminant concentrations in sediment deposits on the Pajarito Plateau are characterized by significant variability as a result of differences in age, particle size, and distance from contaminant source [*Reneau et al.*, 2004]. This variability leads to large uncertainties in the reconstructed ^{137}Cs concentration history (Figure 4), which are especially large for the estimate of peak ^{137}Cs concentration on coarse sediment. However, the ^{137}Cs concentration histories in Figure 4 remain the best available data for computing the input of the tracer to test the model. The impact of these uncertainties on the model results is analyzed in a sensitivity analysis discussed later.

[17] Concentrations were converted to fluxes by multiplying ^{137}Cs concentrations by estimates of the coarse- and fine-sediment discharge from DP Canyon. By integrating empirical sediment transport relations over modeled hydrographs, *Malmon* [2002] computed that DP Canyon contributed an average of 50% (550 T/yr) of the coarse and 80% (900 T/yr) of the fine sediment entering the modeled reaches. Using these values and the curves in Figure 4, an estimated 6,500 mCi of ^{137}Cs entered ULA Canyon from DP Canyon between 1952 and 1997, a large amount compared with the estimated 275 mCi that remained in ULA Canyon in 1997 [*Reneau et al.*, 1998]. In the following sections, the model is used to simulate the movement of

Table 2a. Transition Probabilities for Upper Los Alamos Canyon Model: Coarse Sediment Transition Probability Matrix (Matrix **C**)[a]

	Particle Location at Time t + 1 (*j*)				
Particle Location at Time t (*i*)	Reach 1 Channel	Reach 2 Channel	Reach 3 Channel	Reach 4 Channel	Absorbing State
Reach 1 channel	*0.79*	*0.16*	*0.04*	*0.009*	0.003
Reach 2 channel	*0*	*0.91*	*0.07*	*0.02*	0.005
Reach 3 channel	*0*	*0*	*0.86*	*0.11*	0.03
Reach 4 channel	*0*	*0*	*0*	*0.83*	0.17
Absorbing state	0	0	0	0	1

[a]Transition probabilities are computed using the values in the sediment budget (Table 1) and the equations derived by *Malmon et al.* [2003]. All rows sum to one. Values are annual transition probabilities p_{ij}, defined as the probability that a particle starting in *i* will be in *j* after a single increment of time (1 year). Values in italics are those used to compute the fundamental matrices ($\mathbf{C_B}$ in equation (1)).

this sediment through the 5 km stretch of ULA Canyon from DP to Pueblo Canyon.

4. Particle Trajectories and Sediment Evacuation

[18] The amount of time a particle spends in the valley floor (before entering the absorbing state) is called the transit time for that particle. The expected (mean) transit time for all the particles in a particular deposit is the flushing time of that deposit. *Kelsey et al.* [1987] showed that flushing times can be computed with fundamental matrices. The fundamental matrices for coarse and fine sediment, $\mathbf{S}_C$ and $\mathbf{S}_F$ are

$$\mathbf{S}_C = (\mathbf{I} - \mathbf{C_B})^{-1}$$
$$\mathbf{S}_F = (\mathbf{I} - \mathbf{F_B})^{-1} \quad (1)$$

where $\mathbf{C_B}$ and $\mathbf{F_B}$ are the upper left 4 × 4 submatrices of **C** and **F** (Tables 2a and 2b), consisting of transition probabilities among transient states, and **I** is the identity matrix (a 4x4 matrix in which values along the main diagonal equal one; the remaining entries equal zero).

[19] Calculated flushing times (Tables 3a and 3b) for coarse sediment decrease downstream, from 23 years (for sediment initially stored in Reach 1) to 6 years (for sediment initially in Reach 4). The downstream decrease in flushing time for fine sediment is less pronounced, declining from 63 to 52 years. Flushing times were computed in this way for deposits in Redwood Creek, California [*Kelsey et al.*, 1987]. In that study the flushing times were considerably longer (10^2–10^4 years), which is to be expected because the amount of sediment stored in Redwood Creek is much greater than in ULA Canyon, the reaches are longer, and the return period of dominant geomorphic events is presumably longer.

[20] The matrices $\mathbf{S}_C$ and $\mathbf{S}_F$ (Tables 3a and 3b) contain the average amounts of time that particles will spend in each of the transient states in the valley floor, starting from each of the deposits. However, each deposit contains particles that will follow many different pathways through the valley, so there is a probability distribution of transit times for each deposit. Because these distributions are probably not normally distributed [*Dietrich et al.*, 1982; *Malmon et al.*, 2003] the flushing time (i.e., the mean transit time) may not be a sufficient indicator of residence time for sediment in the valley floor. The probability distributions of particle transit times were computed from the Chapman-Kolmogorov equations, using equations (12)–(13) of *Malmon et al.* [2003]. The transit time distributions for both coarse and fine sediment are strongly right skewed (Figure 5). For coarse sediment the distributions become more strongly skewed in the downstream direction (Figure 5a). For fine sediment, the distributions for all reaches are nearly identical (Figure 5b), reflecting the low probability that fine particles, once mobilized, are redeposited in the floodplain (i.e., low off-diagonal transition probabilities in matrix **F**, Table 2b). The right-skewed nature of the distributions is significant because of the interaction between transit time and the nonlinear process of contaminant degradation (in this case radioactive decay) in determining the mass flux of chemicals out of the system, and the exposure of the humans and other organisms to them during their transit.

[21] It is possible to quantify the timescale over which sediment currently stored within the valley is evacuated and replaced with new sediment from upstream, using the transit time probability distributions (Figure 5) and the masses of the sediment reservoirs [see *Malmon et al.*, 2003, equations (15)–(16)]. The time required for all the particles in the active channel and floodplain to reach the absorbing state

Table 2b. Transition Probabilities for Upper Los Alamos Canyon Model: Fine Sediment Transition Probability Matrix (Matrix **F**)[a]

	Particle Location at Time t + 1 (*j*)				
Particle Location at Time t (*i*)	Reach 1 Floodplain	Reach 2 Floodplain	Reach 3 Floodplain	Reach 4 Floodplain	Absorbing State
Reach 1 floodplain	*0.98*	*0.002*	*0.001*	*0.0008*	0.02
Reach 2 floodplain	*0*	*0.98*	*0.001*	*0.0008*	0.02
Reach 3 floodplain	*0*	*0*	*0.98*	*0.0009*	0.02
Reach 4 floodplain	*0*	*0*	*0*	*0.98*	0.02
Absorbing state	0	0	0	0	1

[a]Transition probabilities are computed using the values in the sediment budget (Table 1) and the equations derived by *Malmon et al.* [2003]. All rows sum to one. Values are annual transition probabilities p_{ij}, defined as the probability that a particle starting in *i* will be in *j* after a single increment of time (1 year). Values in italics are those used to compute the fundamental matrices ($\mathbf{F_B}$ in equation (1)).

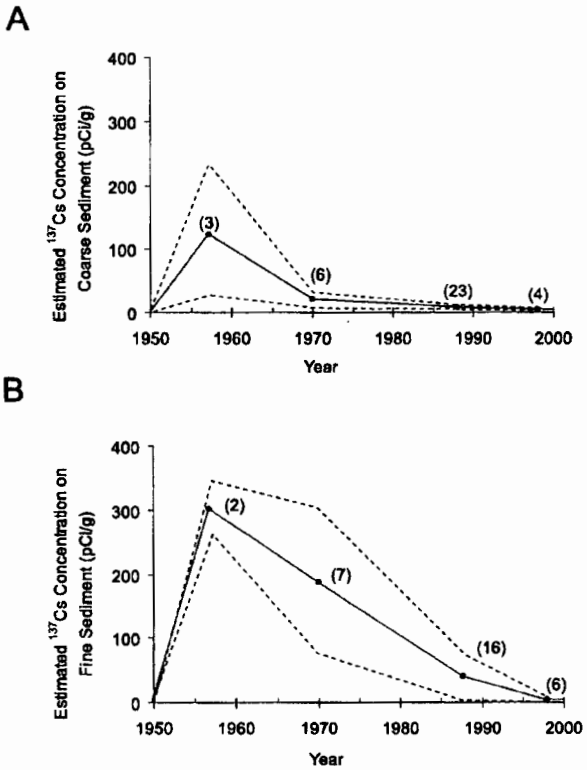

Figure 4. Reconstructed history of ^{137}Cs discharge into the model area at the upstream boundary, based on data compiled by *Malmon* [2002]. Cesium-137 concentration is plotted over time for both (a) coarse sediment and (b) fine sediment. Data points are averages of samples of datable deposits and suspended sediment samples in lower DP Canyon, decay-adjusted to 1997. Numerals in parentheses indicate the number of samples available for each bin. Dashed lines are one standard deviation around the mean concentration values.

approaches infinity, but most of the sediment is evacuated over a timescale of decades. Half the 38,500 metric tons of sediment currently residing in the active part of the 5 km study reach is expected to reach the confluence with Pueblo Canyon in the next 18 years, 90% in 82 years, and 95% in 126 years [*Malmon*, 2002]. This apparently rapid rate of sediment overturn reflects the fact that geomorphic process rates in ULA Canyon are high relative to the mass of sediment stored in active geomorphic units in the valley floor (Table 1). Rates of sediment evacuation are probably several orders of magnitudes longer for lowland rivers with wide valleys (large sediment masses) and low gradients (relatively slower geomorphic process rates).

5. Historical Redistribution of ^{137}Cs

5.1. Cesium-137 Inventory Over Time

[22] The model can track the redistribution of sediment-bound ^{137}Cs in the channels and floodplains of ULA Canyon, by iteratively solving the equation:

$$W(t) = W(t-1)\mathbf{P} + L(t-1) \qquad (2)$$

where $W(t-1)$ is the ^{137}Cs inventory in each reach and in the absorbing state at time $t-1$, $\mathbf{P}$ is the relevant transition probability matrix ($\mathbf{C}$ or $\mathbf{F}$), and $L(t-1)$ is a 1×5 vector containing the amount of ^{137}Cs from DP Canyon that is immediately deposited in each reach and in the absorbing state during the appropriate time step. The sum of $L(t-1)$ is equal to the total amount of ^{137}Cs discharged at the mouth of DP Canyon during the increment of time between $t-1$ and t. The entries in $L(t-1)$ were computed by multiplying the total ^{137}Cs input for the year by the distribution of depositional probabilities downstream of the confluence with DP Canyon (for further details see *Malmon et al.* [2003, equations (19)–(20)]). Radioactive decay of ^{137}Cs was computed at the end of each time step.

[23] The model predicts the rate and timing of contamination and decontamination of the valley from 1950 to 2100. Figure 6 depicts the modeled ^{137}Cs distributions in color for four separate years: (1) 1950, prior to releases at TA-21; (2) 1969, the year when the valley contained the most ^{137}Cs; (3) 1997, the year when the ^{137}Cs distribution was estimated by *Reneau et al.* [1998] and *LANL* [2004]; and (4) 2050, nearly 100 years after the releases began and 65 years after they ceased. Prior to effluent releases at TA-21 (Figure 6a), ^{137}Cs was not present in significant quantities within the floor of ULA Canyon. Although contaminant concentrations on sediment entering ULA Canyon peaked ca. 1957, the modeled peak inventory did not occur until 1969, when approximately 1,000 mCi are thought to have been stored in the valley floor (Figure 6b). By 1997 (Figure 6c), the modeled ^{137}Cs inventories had been reduced to about half this amount, due to the combination of downstream sediment migration and radioactive decay. According to the model, about 95% of the ^{137}Cs present in 1969 will have either left the valley or decayed radioactively by 2050 (Figure 6d).

[24] The modeled histories of ^{137}Cs storage over time reveal a fundamental qualitative difference between the two modes of sediment transport over several decades (Figure 7). Coarse sediment generally moves near the channel bed, and the rate of sediment exchange with the bed is high compared with the downstream flux of coarse sediment (Table 1). While the annual probability that a particle in the bed is mobilized is nearly or equal to one, the probability that it is redeposited in the bed in the same reach is also high. Because of the frequency of sediment exchange with the channel, the ^{137}Cs bound to the coarse sediment moves downstream gradually as a downstream-attenuating wave (Figure 7a). Modeled peak channel ^{137}Cs inventories occur in 1961, 1966, 1969, and 1973, for reaches 1 through 4, respectively.

[25] In contrast, the relatively small fraction of the fine-sediment load that deposits in the floodplain (14%, see Table 1) likely remains there for many decades before being remobilized (the average residence time of a particle in the floodplain prior to being mobilized is approximately 50 years [*Malmon*, 2002]). When floodplain sediment is finally remobilized by bank erosion, there is a relatively small probability (<14%) of being redeposited, either locally or in a downstream reach. In contrast with the gradual downstream migration that characterizes the long-term movement of coarse sediment, the entire floodplain is contaminated and decontaminated simultaneously (Figure 7b). This characteristic

Table 3a. Expected Particle Residence Times in Upper Los Alamos Canyon: Coarse Sediment (Matrix S_C)[a]

Particle Initial Location	Expected Duration in Transient States, years				Flushing Time[b]
	Reach 1 Channel	Reach 2 Channel	Reach 3 Channel	Reach 4 Channel	
Reach 1 channel	5	8	5	4	23
Reach 2 channel	0	11	5	4	21
Reach 3 channel	0	0	7	4	12
Reach 4 channel	0	0	0	6	6

[a]Values computed with the fundamental matrices (see text for explanation).
[b]Flushing time for each storage reservoir is the expected amount of time a particle will remain in the valley floor, starting in the specified location. This is equal to the sum of durations in the transient states. Discrepancies between the sums and the computed flushing times are due to rounding errors.

results from the relatively infrequent exchange of sediment with the floodplain. The amount of contaminant storage in the floodplains over time reflects the balance between the time-varying input of the contaminant from DP Canyon (of which a fraction is deposited overbank), gradual bank erosion of the floodplain, and radioactive decay of the stored ^{137}Cs inventory. The modeled peak ^{137}Cs inventories in the floodplain occur about the same time in all four reaches (1976–1977). Reach 2 stores the greatest amount of ^{137}Cs on fine sediment because it has the widest floodplain [*Malmon*, 2002].

5.2. Modeled and Measured ^{137}Cs Storage in 1997

[26] The model can be tested by comparing the modeled distribution of ^{137}Cs in 1997 with the inventory estimated independently by *Reneau et al.* [1998] and updated in 2003 with new data from previously unsampled reaches. For simplicity, the field estimates are referred to here as "measured" values, with the understanding that they were not directly measured but estimated based on extensive field and laboratory measurements, mapping, averaging, and interpolation. The model was parameterized using a sediment budget and a reconstructed ^{137}Cs input history, while the measured values are based on stratigraphic and geomorphic data from ULA Canyon. Thus the model predictions are being compared with completely independent field data (i.e., the model was not calibrated).

[27] The modeled ^{137}Cs inventory in the ULA Canyon reaches in 1997 is 515 mCi, compared with an estimated inventory from field measurements and sample analyses of about 275 mCi (Table 4). Although the total modeled inventory is 87% higher than the measured inventory, the discrepancy of 240 mCi is less than 4% of the 6500 mCi estimated to have entered the valley from DP Canyon, indicating that the model yields results of the appropriate magnitude. Both the modeled and measured inventories show that the amount of ^{137}Cs stored in the canyon since the late 1990s accounts for only a small portion of the inventory introduced from DP Canyon; the remainder has already left the study reaches or decayed radioactively.

[28] The model reasonably reproduces most of the major spatial patterns of ^{137}Cs storage in ULA Canyon in 1997 (Figure 8). The model predicted that 82% of the contaminant inventory would reside in the floodplain in 1997, compared with 73% of the measured inventory. The longitudinal distribution of ^{137}Cs in the channel in 1997 differs somewhat from the modeled distribution (Figure 8a): the model underpredicted ^{137}Cs inventories in the two upstream reaches and overpredicted them in the downstream reaches. This discrepancy suggests that coarse sediment moves more slowly through the channel than the model predicts, and might reflect an overestimation of the coarse-sediment flux in the original sediment budget, or the observation (discussed earlier) that a fraction of the coarse sediment can be temporarily stored in the floodplain during exceptionally large events or as a result of bioturbation. These mechanisms are not included in the model and would tend to decrease the rate of coarse-sediment migration through the valley floor.

[29] Although the model overpredicted the absolute inventory of ^{137}Cs stored in the floodplain, it replicates the relative longitudinal distribution of storage in the floodplain almost exactly (Figure 8b). This is expected because both the measured and modeled floodplain sedimentation rates (and consequent deposition probabilities) primarily reflect downstream variations in floodplain width, which can be an important variable determining the rate of particle migration through a valley.

Table 3b. Expected Particle Residence Times in Upper Los Alamos Canyon: Fine Sediment (Matrix S_F)[a]

Particle Initial Location	Expected Duration in Transient States, years				Flushing Time[b]
	Reach 1 Floodplain	Reach 2 Floodplain	Reach 3 Floodplain	Reach 4 Floodplain	
Reach 1 floodplain	52	6	3	2	63
Reach 2 floodplain	0	56	3	2	61
Reach 3 floodplain	0	0	53	2	56
Reach 4 floodplain	0	0	0	52	52

[a]Values computed with the fundamental matrices (see text for explanation).
[b]Flushing time for each storage reservoir is the expected amount of time a particle will remain in the valley floor, starting in the specified location. This is equal to the sum of durations in the transient states. Discrepancies between the sums and the computed flushing times are due to rounding errors.

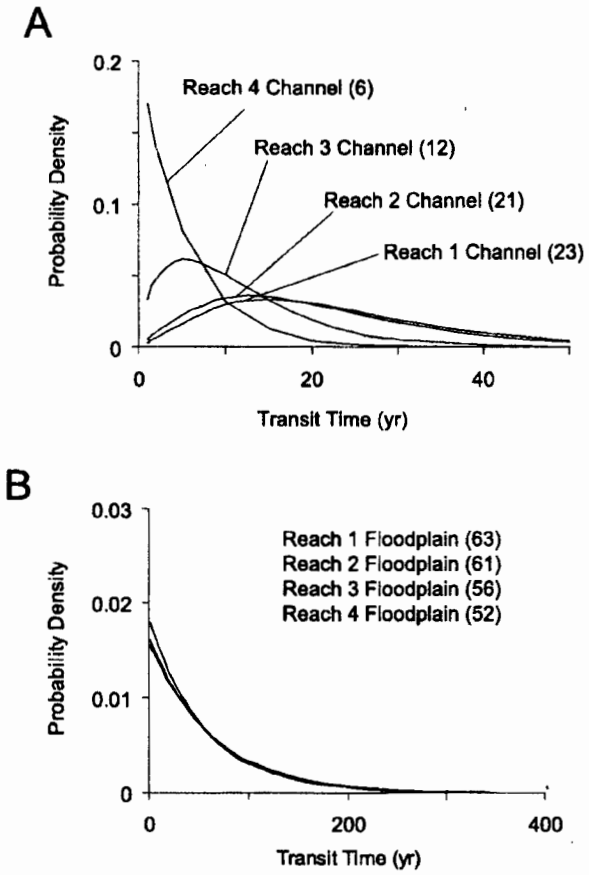

Figure 5. Probability density functions of particle transit time in the valley floor. Transit time is the length of time for a particle to reach the confluence with Pueblo Canyon, starting from a given deposit. Calculated distributions are shown for (a) channel sediment and (b) floodplain sediment residing in each of the four reaches at a point in time. Numerals in parentheses are the mean values of the corresponding probability distributions in years.

5.3. Sensitivity Analysis

[30] In order to better understand how the components of the sediment budget impact the migration of particles and contaminants through the system, and also to explain the source of the discrepancy between the measured and modeled ^{137}Cs inventories, we conducted a sensitivity analysis by repeating the calculations while varying each of the components of the sediment budget over two orders of magnitude. After each repetition, we recorded the total amount of ^{137}Cs predicted to reside within channel and floodplain sediment in the valley floor in 1997, a value that can be compared with the inventories estimated from field data (Figure 9). Note that this method only analyzes the sensitivity of the total predicted ^{137}Cs inventory, and not its longitudinal distribution. The calculations were conducted using the original estimated ^{137}Cs input history (Figure 4, solid lines), then repeated using the mean concentration plus and minus one standard deviation (Figure 4, dashed lines).

[31] The analysis shows that model predictions are significantly affected by the estimated rates of sediment transport and exchange with the floodplain, but that the magnitude of exchange with the channel bed does not strongly impact the calculations (Figure 9). The impact of changing the downstream sediment flux in this steady state model is twofold: on one hand, increasing sediment flux into the system (while keeping the ^{137}Cs concentration constant based on the curves in Figure 4) introduces more of the contaminant into the system from the upstream boundary; on the other hand increasing the fluxes leads to greater rates of sediment migration through the valley. For the coarse sediment (Figure 9a) this leads to a nonmonotonic relationship between sediment flux and predicted inventory, while for the fine sediment (Figure 9b), there is a general increase in the predicted inventory with increased sediment flux. The rate of sediment exchange with the channel bed has a negligible impact because, in a steady state model, increasing the amount of sediment mobilized from the channel bed (and therefore the mobilization probability of coarse particles) will be offset by a higher likelihood that particles are redeposited in the bed within the same reach. In contrast, exchanges with the floodplain have a greater impact on the movement of fine sediment and associated contaminants through the valley. Increasing the rate of floodplain sedimentation significantly increases the probability that suspended particles will deposit somewhere in the valley floor rather than immediately exiting the study reach, leading to higher inventories (Figure 9b).

[32] The sensitivity analysis suggests that an overestimation in the ^{137}Cs concentration on fine sediment entering the upstream end of the study reach is the most likely cause of the discrepancy between the modeled and measured inventories (515 mCi and 275 mCi, respectively). The discrepancy for coarse sediment only accounts for 22 mCi of the difference (Table 4) and could be explained by underestimation of the coarse-sediment flux, overestimation of the ^{137}Cs concentration on coarse sediment entering the reach, or secondary geomorphic processes not included in the model (although accounting for overbank deposition of coarse material and bioturbation, processes observed in the field that deposit coarse sediment in the floodplain, should lead to a further increase in the predicted inventory, not a decrease). In contrast, no reasonable adjustments to the estimated geomorphic process rates could account for the discrepancy between the measured and modeled inventory on fine sediment. The modeled ^{137}Cs inventories computed using the lower bound estimates of the ^{137}Cs input history lie close to measured values (Figure 9b). Thus it is reasonable to hypothesize that the main source of the discrepancies between the modeled and measured inventories is an overestimation of the ^{137}Cs input into the system, an error that is not model-related but results from an inadequate record of contaminant releases.

5.4. Influence of Alluvial Storage on Downstream Contaminant Delivery

[33] One purpose of this paper is to demonstrate how sediment storage in the valley floor modulates the down-

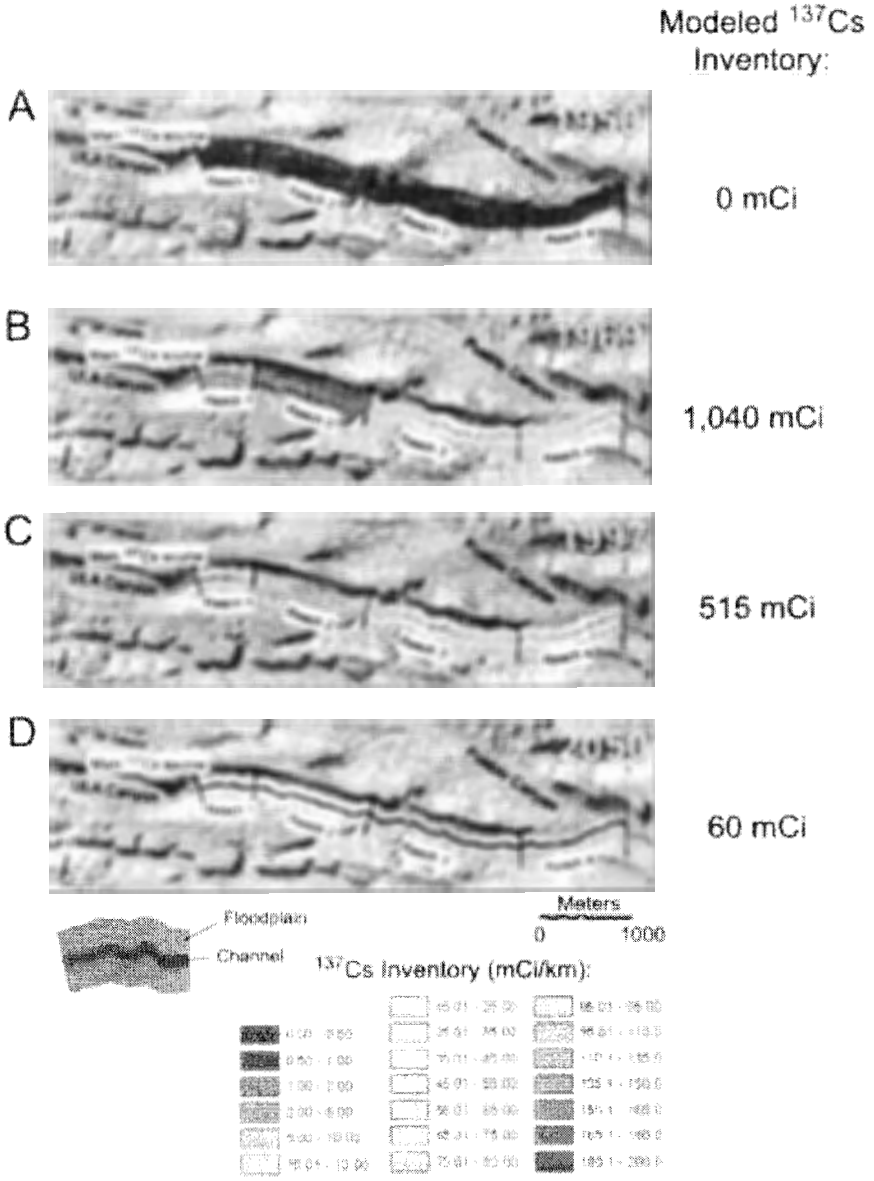

Figure 6. Modeled distribution of ^{137}Cs over time in floodplains and channels in ULA Canyon: (a) in 1950, prior to the first releases; (b) in 1969, when total inventory in the valley floor was greatest; (c) in 1997, when the distribution was estimated independently by *Reneau et al.* [1998] and *LANL* [2004]; and (d) in 2050, after most of the ^{137}Cs has either left the valley or decayed radioactively. The area of active deposition is schematically shown as extending the full width of the canyon bottom, which is wider than the actual extent.

stream delivery of sediment and contaminants. This mechanism can be illustrated by comparing the estimated contaminant fluxes at the upstream boundary with the modeled fluxes at the downstream boundary (Figure 10). For ^{137}Cs associated with coarse sediment (which is exchanged frequently with the channel bed), sediment storage both diffuses the wave of ^{137}Cs that enters the modeled reaches and reduces the total amount delivered downstream, by allowing some of the cesium to decay radioactively in transit (Figure 10a). By contrast, fine-sediment deposition in the floodplain along the 5.3 km reach between DP and Pueblo Canyons amounts to less than 15% of the suspended sediment flux through the valley (Table 1). Therefore most of the ^{137}Cs associated with fine sediment from DP Canyon immediately exited the study reaches (Figure 10b). However, due to the greater affinity of ^{137}Cs for fine sediment, most of the modeled and measured 1997 ^{137}Cs inventories reside in

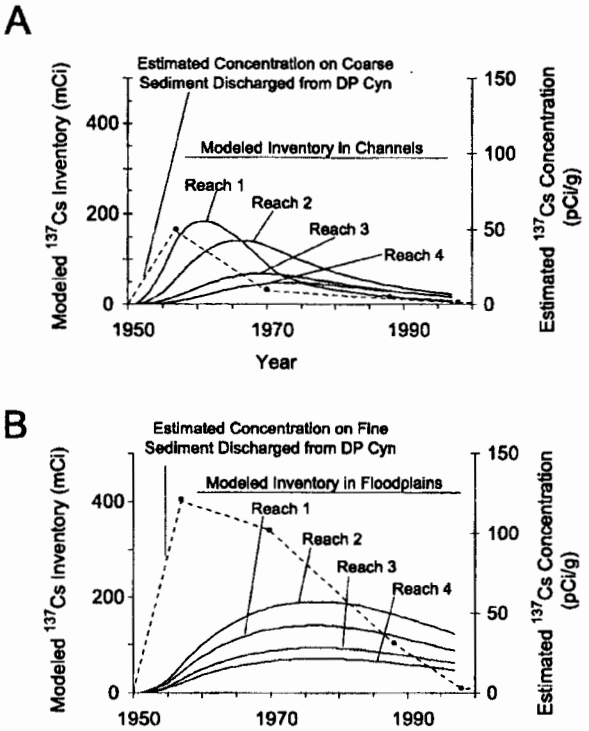

Figure 7. Modeled ^{137}Cs inventory over time in the eight temporary sediment storage reservoirs. (a) Cesium stored in channel deposits; (b) cesium stored in floodplain deposits. The timing of contaminant storage is compared with the timing of contaminant input at the upstream end of the study reach (dashed lines, units on right vertical axis).

the floodplain (note the different vertical scales on Figures 10a and 10b).

6. Future Movement and Delivery of ^{137}Cs

[34] Although most of the ^{137}Cs that entered the study reaches probably exited them on suspended sediment since the 1950s, the future fate of the remaining cesium remains a question. A quantitative prediction of the amount, sources, and timing of future contaminant fluxes in Los Alamos Canyon may be useful for guiding management decisions. Risk models are used to evaluate whether there is an unacceptable risk to human health or ecosystems from ^{137}Cs and other contaminants present on and transported off LANL property, and whether remedial actions are necessary to reduce contaminant transport. Predictions of future ^{137}Cs concentrations and inventories can support these risk assessments and the design of future monitoring programs by quantifying the nature of downstream contaminant remobilization.

6.1. Model Setup

[35] Recent field estimates indicate that about 275 mCi of ^{137}Cs remained in alluvial sediment in 1997, including 73 mCi associated with coarse sediment and 202 mCi on fine sediment (see Table 4, "Measured" columns). This measured ^{137}Cs distribution was adopted as the initial condition for the model predictions in this section. We assumed future ^{137}Cs discharges from DP Canyon (at the upstream end of the model) would decrease linearly from present rates to zero in 2050 [*Malmon*, 2002]. A scenario with linearly decreasing fluxes is the simplest model. The choice of 2050 as the date when concentrations effectively reach zero is considered a conservative estimate, allowing the radioactive decay of 70% of the current ^{137}Cs inventory in DP Canyon and the probable evacuation of much of the remainder.

[36] There are some differences between the spatial extent of mapped inventories and the spatial extent of the model. The model does not separately account for layers of recent fine sediment stored farther from the channel and mapped as f1 and f2 (the fine-grained layers outside the box labeled "Active Sediment" in Figure 2b) by *Reneau et al.* [1998]. These deposits are generally farther from the channel and less susceptible to mobilization than those underlain by recent coarse-facies deposits. These units contain about 20% of the total ^{137}Cs inventory, and are most prevalent in Reach 2. For the sake of simplicity, the model assumes these units have the same probability of being mobilized as the overbank deposits inside the box marked "Active Sediment" in Figure 2b. This approximation will cause some overprediction of the future transport rates of ^{137}Cs associated with fine sediment because residence times in these units are longer than in the units closer to the channel. However, the approximation was necessary as a result of a lack of appropriate field data to compute a separate set of transition probabilities for sediment stored in these deposits.

Table 4. Modeled and Measured Distribution of ^{137}Cs in 1997

	Channel Sediment		Floodplain Sediment		Combined ^{137}Cs Inventory	
	Measured, mCi	Modeled, mCi	Measured, mCi	Modeled, mCi	Measured, mCi	Modeled, mCi
Amount Discharged from Upstream[a]	850		5,700		6,500	
Amount Stored Within the Four Study Reaches[b]						
Reach 1	12	5	32	60	45	65
Reach 2	45	31	118	186	163	217
Reach 3	10	27	37	101	47	128
Reach 4	6	32	16	74	21	106
Total ^{137}Cs inventory in study area in 1997	73	95	202	420	275	515
Amount stored downstream of study area		310		2,500		2,800
Amount radioactively decayed as of 1997		440		2,800		3,200

[a]Estimated amount of ^{137}Cs discharged from DP Canyon into the study reaches; values computed by integrating under the solid lines in Figure 4.
[b]Measured and modeled distributions of ^{137}Cs in 1997 within the study reaches.

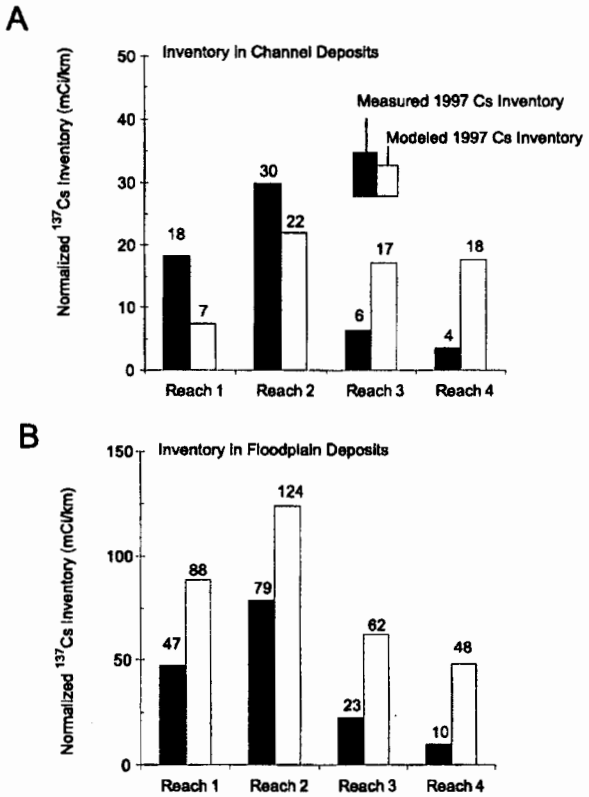

Figure 8. Comparison of measured and modeled ^{137}Cs distribution in 1997. (a) Inventories in the channel bed. (b) Inventories in the floodplain. The inventories are normalized by reach length (mCi/km) to display downstream variations in the amount of ^{137}Cs storage.

[37] Model predictions in this section account for the removal in summer 2000 of a small deposit of relatively highly contaminated sediment near the mouth of DP Canyon. The impact of this sediment removal on downstream ^{137}Cs transport rates is discussed further below.

6.2. Future Sources and Transport of ^{137}Cs

[38] Assuming historical sediment transport conditions prevail (i.e., using the same probability matrices **C** and **F** that were used in previous sections), the model predicts that more than 50% of the 275 mCi of ^{137}Cs associated with active sediment in the valley floor in 1997 will decay radioactively before reaching the confluence with Pueblo Canyon (Figure 11). Presently, the channel and floodplain contribute nearly equally to the modeled downstream contaminant flux. After the wave of ^{137}Cs-contaminated coarse sediment has passed the downstream model boundary (about 2030), sediment currently stored in floodplains will contribute nearly all the ULA-derived ^{137}Cs flux near the property boundary (Figure 11).

[39] The model predicts that in the next half century, 60 to 80% of the ^{137}Cs flux at the downstream boundary will be transported on fine sediment (Figure 12a). Coarse sediment is predicted to transport a relatively higher proportion of the total ^{137}Cs flux over the next two decades (up to 35% in 2008, Figure 11), but fine sediment dominates long-term contaminant discharge (Figure 12a).

[40] The calculations provide a mathematical decomposition of the future contaminant efflux from the study reach, and allow us to compare the relative contributions of various upstream sources to downstream contaminant fluxes. Sediment initially stored in the channels, the floodplains, and in DP Canyon are predicted to each account for about a third of the ^{137}Cs discharge from ULA Canyon over the next decade. Over several decades, however, the floodplains will contribute much more than the channels to contaminant discharges (Figure 12b). The calculations can also provide a spatial picture of future contaminant sources: the model predicts that most of the ^{137}Cs flux over the next half century will originate in Reach 2 and in DP Canyon (Figure 12c). Reach 2 initially contains the greatest amount of ^{137}Cs, and therefore contributes the most to downstream contaminant fluxes over the next 20 years. After that, ^{137}Cs currently stored in DP Canyon is predicted to dominate the contaminant efflux at the downstream boundary.

6.3. Management Implications of Model Predictions

[41] In many management scenarios involving contaminated sediment, a primary objective is to reduce contaminant fluxes or concentrations at a particular downstream boundary. In such cases, probabilistic calculations of sediment redistribution could support decision making. In May 2000, the Cerro Grande fire burned the headwaters of many of the canyons that cross the Pajarito Plateau, increasing the frequency and magnitude of floods through LANL property [*Shaull et al.*, 2004]. The potential flood threat raised concerns that high flows would cause significant erosion and downstream transport of contaminated sediment. These concerns motivated two remediation measures in ULA Canyon in summer 2000 to reduce ^{137}Cs discharges at the LANL property boundary: (1) excavation of a small deposit of relatively highly contaminated sediment in ULA Canyon just below the DP Canyon confluence; and (2) construction of a porous rock-gabion barrier ("low-head weir") and an upstream settling basin to trap sediment in the lower portion of ULA a short distance upstream of the Pueblo Canyon confluence (Figure 1). The model provides a tool that can be used to quantify the potential long-term effects of these projects and support recommendations for management strategies based on model predictions of future contaminant transport.

[42] The excavation project removed about 440 m^3 of sediment located just downstream of the mouth of DP Canyon. The excavation targeted deposits that contained the highest measured ^{137}Cs concentrations in ULA Canyon, and that were also susceptible to erosion by floods. The excavated site contained approximately 14 mCi of ^{137}Cs [*Reneau et al.*, 1998], or 5% of the estimated total ^{137}Cs inventory in the valley floor, concentrated along a 50 m length of channel in modeling reach 1. We modeled the excavation by reducing the initial ^{137}Cs inventory in the reach 1 channel and floodplain by 5 and 9 mCi, respectively, and repeating the calculations. The modeled impact of the excavation on ^{137}Cs concentrations and fluxes near the LANL boundary was minor (Figure 13). This example shows how the model can provide a quantitative prediction of the poten-

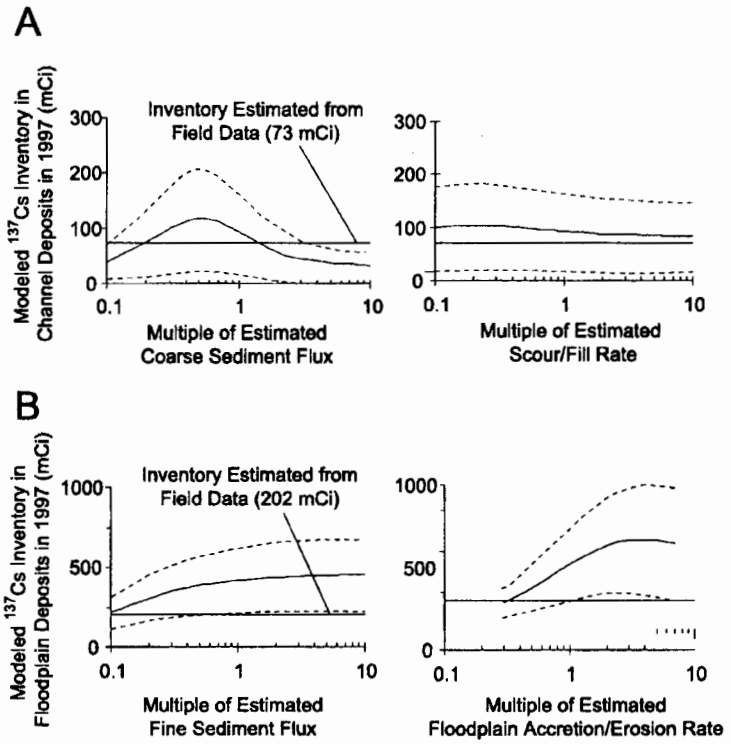

Figure 9. Sensitivity of modeled ^{137}Cs inventory in 1997 to the components of the sediment budget and the estimation of the ^{137}Cs input to the model system at the upstream boundary. (a) Sensitivity to variations in coarse-sediment flux and the rate of sediment exchange with the bed. (b) Sensitivity to variations in fine-sediment flux and the rate of sediment exchange with the floodplain. Dashed lines are model results computed using the estimated ^{137}Cs concentration on incoming sediment, plus and minus one standard deviation (see Figure 4). The corresponding measured inventories are noted on each plot with a horizontal line.

tial effects of upstream remediation actions (such as the excavation) on contaminant delivery to downstream areas.

[43] The low-head weir was designed to capture primarily the gravel and sand fractions of the load during a calculated postfire, 100-year flood. However, fire-related flooding in ULA Canyon was small compared with that in other watersheds, largely because floods initiated in the upper burned portions of the watershed were dissipated in the Los Alamos Reservoir (Figure 1), which lies below most of the burned area. Figure 11 shows that most of the ^{137}Cs load in the next half century will be carried by fine sediment, and a long-term sediment budget [*Malmon et al.*, 2004] indicates that most of this will probably be transported by small to moderate floods. The efficiency of the weir to trap finer-grained sediment and associated ^{137}Cs in small and moderate floods is not known. However, the analyses suggest that the effectiveness of a sediment trapping structure as a measure to reduce long-term contaminant transport might have been maximized by focusing the design on deposition of the finer-grained fraction during relatively small floods, rather than the coarse fraction of very large floods. Though these modeling results were not available prior to construction of the weir, this example shows how simple probabilistic calculations based on a sediment budget (specifically the ability to mathematically decompose the future contaminant load) could be used to aid in the engineering design of mitigation measures.

[44] According to these calculations, a long-term strategy to reduce contaminant discharge from the study reaches should emphasize solutions that reduce transport of the fine component of the sediment load, particularly during relatively small events (i.e., those with recurrence intervals of $\leq$2 years). The calculations support remediation strategies that reduce the probability of floodplain-stored contaminated sediment being mobilized in small to moderate floods, or measures that enhance its redeposition downstream. In addition to alternatives involving sediment removal or engineered structures, bank stabilization and vegetation enhancement strategies may be appropriate in some settings. Because the primary goal of such activity would be to reduce floodplain erosion and increase sediment deposition during small to moderate floods (rather than protecting against extremely large floods), the goal could possibly be achieved with relatively inexpensive and nonintrusive actions. Probabilistic analysis of particle trajectories such as the example presented in this paper would have utility in designing and targeting areas for such work.

7. Discussion and Conclusions

[45] Contaminant fate in the fluvial environment is determined by the relative timescales of contaminant degradation and long-term sediment movement through

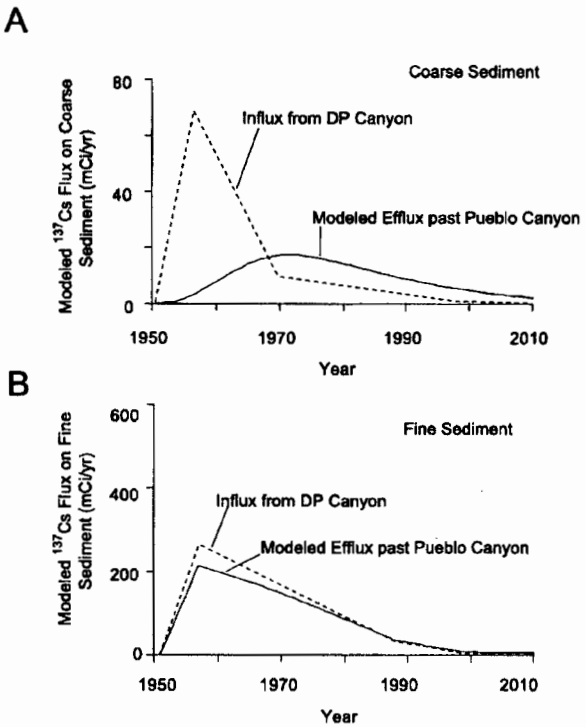

Figure 10. Estimated ^{137}Cs influxes (see Figure 4) and modeled ^{137}Cs effluxes from the study reach over time. (a) Contaminant flux carried by coarse sediment. (b) Contaminant flux carried by fine sediment.

the sediment reservoirs in the valley floor. The theory formalized by *Malmon et al.* [2003] computes sediment trajectories through alluvial sediment storage reservoirs, and provides a convenient framework for analyzing the fate of contaminated sediment in valleys. This paper tested that theory in a field setting, using sediment-bound ^{137}Cs as a sediment tracer over the past 50 years.

[46] In our field area the approach realistically reproduced the approximate current magnitude and distribution of ^{137}Cs in the study reach from an estimate of the influx from upstream. The model illustrates how channel and floodplain storage modulate ^{137}Cs delivery from upper Los Alamos Canyon, and provides insight into the long-term migration of sediment and contaminants through a small, alluvial valley in a semiarid environment. The portion of the contaminant load associated with the coarser bed material sediment (generally sand particles larger than 0.25 mm diameter) is exchanged frequently and for relatively short durations with the channel bed; as a result the contaminant pulse on coarse sediment moves through the valley as a downstream-attenuating wave. Frequent storage of coarse sediment allows the contaminant wave to diffuse and much of the ^{137}Cs to radioactively decay before reaching the downstream boundary.

[47] By contrast, most of the ^{137}Cs associated with fine sediment passes through the valley within hours after entering it from DP Canyon; only 14% of the fine sediment in floods is predicted to be deposited on the floodplain within the study reach. However, these particles will be gradually eroded from floodplain storage over periods of decades, making the floodplain a long-term contaminant source. The amount of ^{137}Cs in the floodplain at a given time reflects the balance between the time-varying input of the contaminant from DP Canyon, gradual bank erosion of the floodplain, and radioactive decay of the stored ^{137}Cs inventory. Most of the ^{137}Cs that entered the study area has already passed through the 5 km study reach. Of the current inventory, approximately half is predicted to radioactively decay before reaching the downstream model boundary.

[48] While the theory works well in upper Los Alamos Canyon, it may not be immediately transferable to many other settings in the form outlined by *Malmon et al.* [2003], who point out that the equations they developed simplify or ignore several important aspects of sediment routing in river valleys, including (1) the influence of particle size sorting and selective transport; (2) nonsteady state conditions; and (3) the stochastic nature of forcing mechanisms (as opposed to the stochastic nature of particle trajectories, which the theory does represent explicitly).

[49] The model reported in this paper differentiated between two particle size classes, coarse and fine sediment, following the observation [*Malmon et al.*, 2004] that these two populations are distinct with respect to transport mechanisms, storage reservoirs, and contaminant concentrations. The two particle sizes were modeled by creating two separate probability models and parameterizing them with

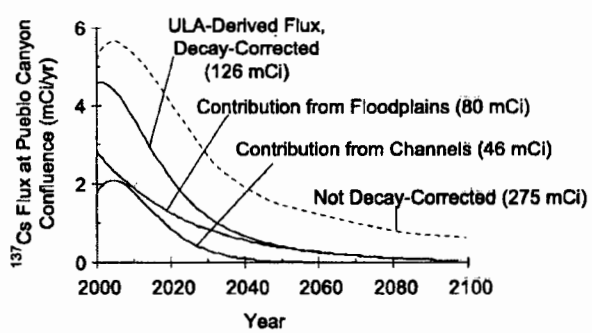

Figure 11. Modeled future fluxes of ^{137}Cs at the downstream boundary (the confluence with Pueblo Canyon) derived from sediment stored in ULA Canyon in 1997. Transport of ^{137}Cs discharged from DP Canyon after 1997 is not included in this graph. Plots show the fluxes of ^{137}Cs into the absorbing state associated with particles identified by their location at the beginning of the model run. The initial condition for this model run was the spatial distribution estimated in 1997 [*Reneau et al.*, 1998; *LANL*, 2004] and accounts for the excavation and removal of a small, relatively highly contaminated deposit near the mouth of DP Canyon in summer 2000. Numerals in parentheses indicate the total amount of the original ^{137}Cs inventory expected to reach the downstream boundary beginning from the channels and floodplains along the study reach. Dashed curve represents fluxes that would occur if ^{137}Cs did not undergo radioactive decay, illustrating that temporary sediment storage allows more than 50% of the contaminant to decay before reaching the downstream boundary.

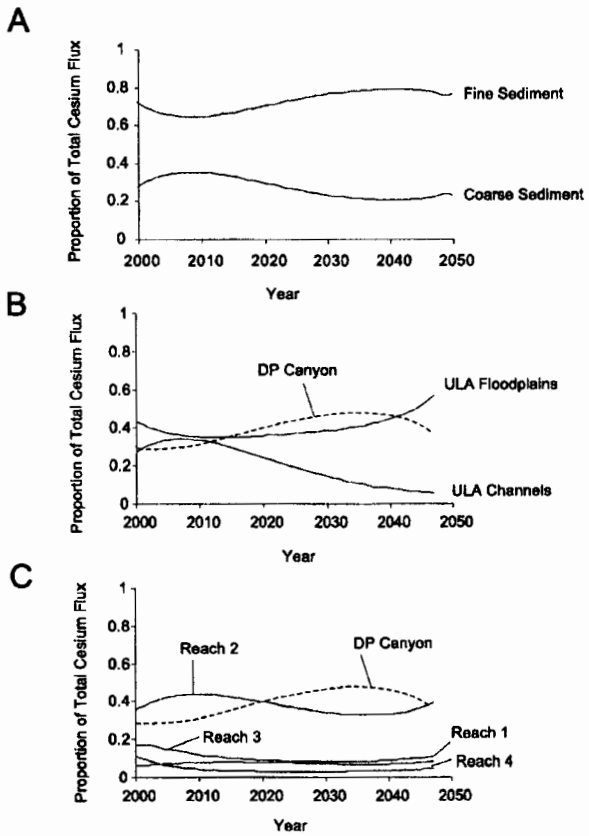

Figure 12. Decomposition of the contaminant load at the downstream boundary. Plots show model predictions of the relative amount of the ^{137}Cs load over time contributed by various types of upstream sources. (a) Relative contributions of coarse versus fine sediment to the future ^{137}Cs load. (b) Relative contributions of channels and floodplains along ULA Canyon and of DP Canyon to the future ^{137}Cs load. Each line indicates the proportion of the contaminant load over time contributed by sediment initially stored in each of the indicated locations. (c) Relative contributions of the four model reaches and DP Canyon to the future ^{137}Cs load.

separate sediment budgets for the coarse- and fine-grained fractions. This is an advance over the single-fraction model outlined by *Malmon et al.* [2003] and appropriate to the local conditions. However, a two fraction model may not be the ideal approach for other applications, for example where only a fine fraction is of concern (in which case a single particle size model would be sufficient), or in a study of the long-term bed load movement, sorting, and abrasion in gravel bed rivers (where more than two size fractions may be required).

[50] Upper Los Alamos Canyon has remained relatively stable over the past 50 years, which led to an assumption of steady state conditions that greatly simplified the application of the model and reduced the necessary input data requirements. Similar assumptions can be made for many, but not all, alluvial valleys over timescales relevant to the long-term migration of sediment and associated constituents. For example, in the case of a release that introduces contaminants to a system but does not significantly change the mass of sediment moving through a valley, the model should be transferable to the extent that a reliable sediment budget can be estimated. However, the theory remains untested for nonsteady state field conditions. Such conditions are characteristic of many problems involving the fate of large volumes of contaminated material into rivers, such as following tailings dam breaches or dam removal projects. Future field applications would help test and refine the theory for nonsteady state conditions.

[51] The current application employed a simplified model in which the transition probabilities remained constant from year to year. In reality, even in systems that remain in steady state, the sediment budget is driven by events that are themselves characterized by significant temporal variability. Locally, the sediment budget is dominated by relatively low return period events [*Malmon et al.*, 2004], so over several decades using a single transition probability matrix should not impact the model predictions. However, in settings where the return periods of dominant geomorphic events are long compared with the timescale of interest, the migration of sediment and contaminants may depend on how many times a particular type of event occurs within the time frame represented by the model. *Malmon et al.* [2003] propose a possible approach to incorporating the stochastic nature of forcing events into the probability framework, and other strategies may also be valid. Such elaborations could be useful for quantifying the probabilities of various outcomes in places where the sediment budget is dominated by high magnitude, low-frequency geomorphic events.

[52] Sediment and contaminant delivery from watersheds can be strongly influenced by sediment exchanges within

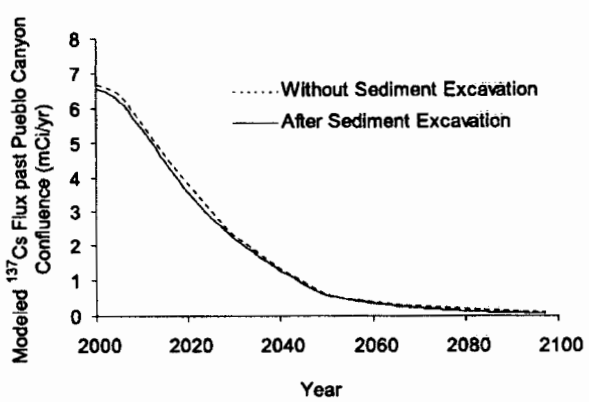

Figure 13. Modeled impact of sediment excavation undertaken in summer 2000. About 440 m^3 of ^{137}Cs-contaminated sediment was removed from a deposit near the upstream end of reach 1. The deposit contained an estimated inventory of 14 mCi. The scenario was modeled by reducing the initial inventory in the reach 1 channel by 5 mCi and the floodplain by 9 mCi. The model predicts that excavation of 14 mCi of ^{137}Cs near the mouth of DP Canyon will reduce the total ^{137}Cs delivery by 6 mCi. The remaining 8 mCi would have decayed radioactively in temporary storage.

the valley floor. Migration of sediment and associated contaminants through alluvial valleys is controlled by the rates of sediment transport, deposition, and erosion, and by the masses of the sediment reservoirs on which these processes operate. In our study area the annual rates of geomorphic processes are large compared with the amount of active sediment stored in the valley, so the timescale of sediment overturn is short, on the order of 10^1-10^2 years. In lowland river valleys, which typically store more sediment, have lower gradients, and respond to seasonal signals rather than discrete events, the rate of sediment overturn should be much slower.

[53] In general, particles and associated pollutants enter temporary sediment storage reservoirs such as channels, floodplains, and river deltas. The fate of sediment-bound contamination depends on the frequency of sediment exchange with and duration of storage in such reservoirs. Probabilistic analysis of particle trajectories provides a realistic approach for quantifying these mechanisms, and a useful platform for managing contaminated sediment in many alluvial river valleys.

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