Pulsed Redistribution of a Contaminant Following Forest Fire: Cesium-137 in Runoff

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ABSTRACT

Of the natural processes that concentrate dispersed environmental contaminants, landscape fire stands out as having potential to rapidly concentrate contaminants and accelerate their redistribution. This study used rainfall simulation methods to quantify changes in concentration of a widely dispersed environmental contaminant (global fallout ^{137}Cs) in soils and surface water runoff following a major forest fire at Los Alamos, New Mexico, USA. The ^{137}Cs concentrations at the ground surface increased up to 40 times higher in ash deposits and three times higher for the topmost 50 mm of soil compared with pre-fire soils. Average redistribution rates were about one order of magnitude greater for burned plots, 5.96 KBq ha^{-1} mm^{-1} rainfall, compared with unburned plots, 0.55 KBq ha^{-1} mm^{-1} rainfall. The greatest surface water transport of ^{137}Cs, 11.6 KBq ha^{-1} mm^{-1}, occurred at the plot with the greatest amount of ground cover removal (80% bare soil) following fire. Concentration increases of ^{137}Cs occurred during surface water erosion, resulting in enrichment of ^{137}Cs levels in sediments by factors of 1.4 to 2.9 compared with parent soils. The elevated concentrations in runoff declined rapidly with time and cumulative precipitation occurrence and approached pre-fire levels after approximately 240 mm of rainfall. Our results provide evidence of order-of-magnitude concentration increases of a fallout radionuclide as a result of forest fire and rapid transport of radionuclides following fire that may have important implications for a wide range of geophysical, ecosystem, fire management, and risk-based issues.

Certain processes, such as bioaccumulation, are known to concentrate dispersed environmental nutrients and contaminants. Such concentration processes stand out in contrast to the many natural processes that decrease concentration levels, such as dilution, dispersion, chemical change, and radioactive decay (Whicker and Schultz, 1982; Kendall and McDonnell, 1998; Peles et al., 2000). Of the processes that concentrate, those associated with wildfire can occur rapidly and are thought to contribute to landscape-scale redistribution of nutrients and contaminants (Paliouris et al., 1995; DeBano et al., 1998). Examination of fire as an agent in concentrating and redistributing environmental contaminants, carbon, and nutrients has relevance to a wide range of current interests including ecosystem dynamics (Baird et al., 1999; Thomas et al., 1999; Breshears and Allen, 2002); forest burning as a land use practice (Kauffman et al., 1993); increased potential for landscape fires from climate change and forest fuel buildup (Piñol et al., 1998; Mast et al., 1999; Moore et al., 1999; Watson and The Core Writing Team, 2001); and mobility of dispersed radionuclides (Kashparov et al., 2000; Johansen et al., 2001b; Whicker et al., 2002). Better information on fire-induced concentration and redistribution of contaminants may be particularly useful in assessing risks and doses to human and environmental receptors. Routine risk assessments, and the models used for these assessments, may underpredict risks over long time frames when they do not include concentration and redistribution from periodic events such as fire (Whicker et al., 1999).

Quantifying the processes that concentrate and redistribute nutrients has been accomplished through use of tracers, including fallout radionuclides such as ^{137}Cs, which are widely dispersed in world ecosystems (Kendall and McDonnell, 1998; van der Perk et al., 2002). However, few such measurements have been made following landscape-scale fires. In a Canadian boreal forest, elevated concentrations of ^{137}Cs were found in surface soils at an area that had been burned years before, indicating redistribution from burned aboveground matter to the ground surface and concentration of ^{137}Cs in deposited ash (Paliouris et al., 1995). Amiro et al. (1996) burned various vegetation types and found that on a unit weight basis, the ash deposits after fire were enriched in elemental cesium from 4 to 25 times during field burns, and more than two orders of magnitude in laboratory burns compared with the original vegetation. These studies suggest that following wildfire, potentially large increases may occur in the concentration levels of fallout radionuclides in ash and soils at the ground surface.

Subsequent to deposition of concentrated radionuclides on the ground surface, erosion and transport of concentrated radionuclides by surface water runoff are expected to occur. In the Paliouris et al. (1995) study, the total ^{137}Cs inventory of the burned area (total in soils and biomass) was less than that of an unburned control area, indicating redistribution away from burned areas. The authors attributed ^{137}Cs losses from the burned area to volatilization and fly-ash processes during fire, and surface water runoff after fire. In the same study, ^{137}Cs inventories in both burned and unburned areas were highest at the ground surface (up to 55 KBq ha^{-1} in the organic soil layer) compared with the lesser aboveground inventory (up to 14 KBq ha^{-1}) further suggesting the importance of surface water erosion in redistributing ^{137}Cs. However, these inventory measurements were made years after fire occurrence, thus considerable uncertainty remains regarding rates of concentration and redistribution, and whether accelerated erosion and transport of fallout radionuclides by surface water can occur on post-fire landscapes.

Concentration changes of ^{137}Cs during rainfall and runoff after controlled fire in grassland and shrubland

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were studied by Johansen et al. (2001b), who found that runoff from burned plots yielded up to 22 times greater amounts of ^{137}Cs compared with unburned plots (maximum yields of up to 0.28 KBq ha^{-1} mm^{-1} rainfall). Most of the increased ^{137}Cs transport was associated with the amplified sediment transport from burned surfaces. Little ash was observed to remain after the controlled fires on the grassland and shrubland plots.

Part of the increase in ^{137}Cs transport from burned plots observed by Johansen et al. (2001b) was attributed to enrichment of ^{137}Cs in runoff. Enrichment can occur during erosion and runoff through preferential entrainment of fine particles such as clay-sized particles, which have greater affinity for cationic contaminants such as ^{137}Cs (Graf, 1971; Menzel, 1980; Lane and Hakonson, 1982; Watters et al., 1983; Lane et al., 1986; Hakonson and Lane, 1993; Weigand et al., 1998). This enrichment effect appears to occur on both unburned and burned landscapes; however, measurements are lacking for the enrichment effect following landscape fires where soil can reach high temperatures and thus potentially affect enrichment processes by changing soil characteristics.

These earlier studies suggest a series of processes during and after fire consisting of ashing of biomass, amplified surface water erosion, enrichment during surface water erosion, and accelerated transport in runoff that result in concentration of fallout radionuclides and subsequent rapid redistribution away from burned areas. However, data are lacking that quantify these processes after a major landscape fire. In particular, measurements are lacking that relate specific fire effects, such as removal of vegetation, to contaminant transport.

Our primary objective was to quantify changes in the concentrations of a global nuclear fallout radionuclide (i.e., ^{137}Cs) at the ground surface and in surface water runoff following a major forest fire. Specifically, we used data from rainfall simulation plots to evaluate post-fire changes in (i) ^{137}Cs concentrations in soils, (ii) ^{137}Cs concentrations and redistribution rates in surface water runoff (both changes in concentration and duration of those changes), and (iii) associated changes in the enrichment of ^{137}Cs in transported sediments compared with their parent soils. In addition, we compared ^{137}Cs transport from rainfall simulations in forest with transport from prior rainfall simulations in shrubland and grassland ecosystems. This study was conducted to provide insight into the underlying processes governing post-fire movement of environmental contaminants and to provide results relevant to a wide range of geomorphic, ecosystem dynamics, risk, and fire management applications.

MATERIALS AND METHODS

Study Site

The study site is located within the Pajarito Watershed near Los Alamos, New Mexico, in the southwestern USA (Fig. 1). The watershed has a semiarid, temperate mountain climate, with an average annual precipitation of about 500 mm, with the major portion occurring in July and August (Bowen, 1990). Study plots have loam soils, consisting of about 40% sand, 47% silt, and 13% clay (Johansen et al., 2001a). Site soils were identified as Typic Eutroboralfs by Nyhan et al. (1978). Site vegetation is dominated by mature ponderosa pine (*Pinus ponderosa* C. Lawson) and Gambel oak (*Quercus gambelii* Nutt.).

In May 2000, the Cerro Grande wildfire burned approximately 17 400 ha in the Los Alamos area. Rainfall simulation plots were established about 11 wk later at burned and unburned locations approximately 150 m apart within the Pajarito Watershed, near the existing Two Mile Mesa meteorological station operated by the Los Alamos National Laboratory (35°51′49″ N, 106°19′42″ W). Fire occurred at the burned plots site on 10 and 11 May 2000. During burning, all grasses and forbs were consumed and most Gambel oak was burned with some residual stalks remaining. The needles and small limbs of the ponderosa pine trees were consumed to the full height of the trees (about 15 m). The post-fire ground surface was loose soil overlain in some areas by light-colored ash deposits varying in thickness from 0 to approximately 20 mm. Before the Cerro Grande fire, a duff layer existed at the burned plot site ranging from 0 to about 40 mm deep. This layer is thought to have remained unburned for at least the prior 60 years

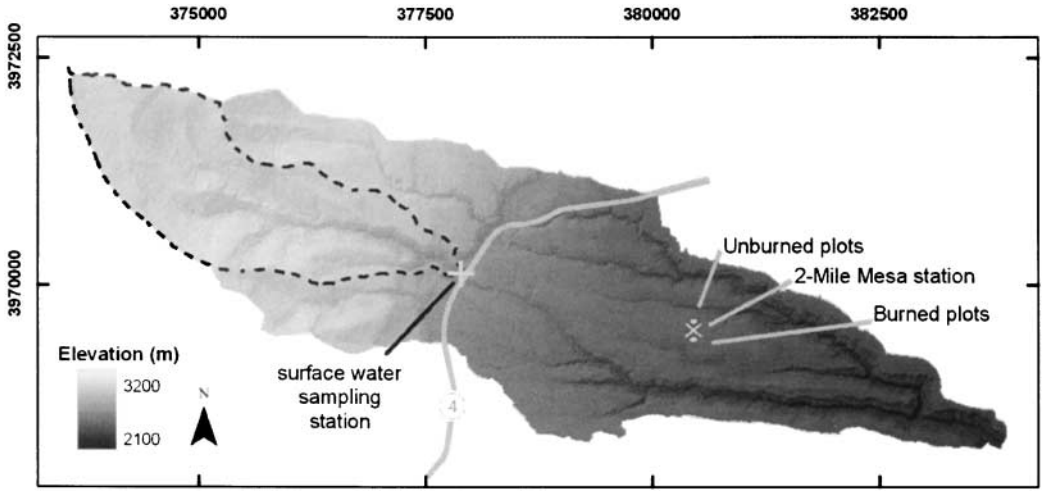

Fig. 1. Study locations are shown within the Pajarito Watershed (shaded) including the approximately 480-ha upper watershed area (dashed line) that contributed to stormwater runoff measurements. New Mexico State Road 4 is shown. The community of Los Alamos, New Mexico is located approximately 3 km northeast of the study area.

and potentially earlier (Touchan et al., 1996) and was thus available to accumulate atmospheric fallout ^{137}Cs resulting from the open-air testing of nuclear weapons that began in the 1940s and peaked in the late 1950s and early 1960s.

This study site was selected in part for its location within the Los Alamos National Laboratory where a baseline of environmental radionuclide data existed before the Cerro Grande wildfire. Pre- and post-fire concentrations of ^{137}Cs in soil were available from annual sampling at a location at the midpoint between the unburned and burned study plots (Two Mile Mesa site; Los Alamos National Laboratory, 2001). Ash sampling following the fire was conducted by the New Mexico Environment Department (personal communication, 2001) at the burned study plot site and other nearby locations within the Pajarito Watershed. Pre- and post-fire concentrations of ^{137}Cs in watershed stormwater runoff were available from sampling at the nearest surface water sampling station (#E240) approximately 2 km west of the study plots (Los Alamos National Laboratory, 2001). This station collects runoff samples from the approximately 480-ha upper area of the Pajarito Watershed that has an average slope of about 8%. Following the Cerro Grande fire, about 52% of the Pajarito Watershed area was estimated to have burned to a high-severity condition and about 44% to a low-severity condition as qualitatively defined by a Burned Area Emergency Rehabilitation team (USDA Forest Service, 2000). In addition, ^{137}Cs concentrations in vegetation at the study plot site were determined before wildfire through sampling of ponderosa tree-shoot tips and composited grass samples (Gonzales et al., 2000). The hydrology in ponderosa pine forest within the Pajarito Watershed has been extensively characterized with respect to surface water runoff and subsurface interflow (Wilcox et al., 1997; Newman et al., 1998; Wilcox and Breshears, 1998), soil water dynamics (Brandes and Wilcox, 2000), and infiltration (Newman et al., 1997). Related Cerro Grande fire runoff, erosion, and sediment yield studies at nearby similar locations include Beeson et al. (2001), Cannon et al. (2001), Malmon et al. (2002), McLin et al. (2001), and Wilson et al. (2001).

Experimental Design

Measurements of ^{137}Cs concentrations in runoff were made from 12 surface water runoff events that were generated using the rainfall simulation methods reported by Johansen et al. (2001a). Briefly, four rainfall simulation plots, each 3.03 by 10.7 m (10 by 35 ft), were established; two in the wildfire-burned area and two control plots at a location approximately 150 m away in an unburned area. The percentages of ground cover (including rock, charred wood, and vegetation) on burned and unburned plots were characterized by point frame analysis (Levy and Madden, 1933). In addition, 48 soil samples of the topmost 50 mm of soil were gathered at the study plots and texture analysis was performed using sieving and pipette analysis (using American Society for Testing and Materials methods referenced in Klute, 1986). In all cases, the soil sampled was the topmost 50 mm of soil. If ground litter was present, it was removed before collecting soil samples. Because of the unforeseen nature of wildfire, the plots could not be paired with identical characteristics. However, the soil series, plot dimensions, rainfall intensities, and amount of rainfall received were similar for the burned and unburned plots. The plots did vary in slope with averages of 4.5% for unburned plots and 7.0% for burned plots. These differences in slope are unlikely to make a large difference in sediment yields (Wilcox et al., 1988).

Rainfall simulation was used to generate runoff in lieu of natural storms to provide for control of rainfall amounts and intensities. Rainfall simulations were initiated 31 July 2000, 80 d following burning at the study site by the Cerro Grande fire. During the interval between burning and experimentation, approximately 51 mm of precipitation occurred, primarily in one storm event on 28 June 2000. During the study, a 16-m-diameter, rotating-boom rainfall simulator applied rainfall at a rate of 60 mm h^{-1} in three separate runs that totaled about 120 mm of applied rain. The three runs consisted of a 1-h run (labeled "dry run" for its antecedent moisture condition), followed by a 24-h interval of no rain, then second and third rainfall events ("wet" and "very wet runs") separated by a 0.5-h period of no rain. A 1-h storm at 60 mm h^{-1} represents approximately a 100-yr recurrence interval at Los Alamos. This study design is consistent with and thus provides for comparison with previous research (Renard, 1986) including that performed by the USDA Agricultural Research Service. The drop-size distribution from the rainfall simulator nozzles was similar to that from natural rainfall, but the drops impacted the ground surface with about 80% of the kinetic energy of natural rain (Swanson, 1965). One-liter samples of unfiltered runoff (water and sediment) were collected every 2 to 4 min at the downslope end of each plot in a gutter system that channeled runoff through a calibrated flume where flow measurements were made using a bubble gage flow meter (ISCO, Lincoln, NE). The runoff samples were stored in polyethylene bottles within sealed boxes in an air-conditioned laboratory for more than 24 d during which the sediment fraction settled. A 5-mL sample was taken of the water fraction to test for dissolved and colloidal-associated ^{137}Cs. The remaining approximately 1-L sample was then decanted, the sediments dried and weighed, and a portion of the sediment was wet-sieved to determine grain-size distributions.

In addition to soil and water samples, additional samples were gathered of the litter decomposition layer that exists between loose litter and mineral soils in ponderosa pine forest at Los Alamos Canyon, about 2 km from the study plots, and at three additional ponderosa pine forest locations in the western USA (Gila National Forest, New Mexico, 33°05′38″ N, 108°11′12″ W; La Veta Pass, Colorado, 37°37′03″ N, 105°13′49″ W; and near Fort Collins, Colorado, 40°35′38″ N, 105°22′50″ W). At each location, three samples were gathered from locations at least 0.5 km apart in mature ponderosa forest areas where no evidence of fire was visible. At Los Alamos, the Los Alamos Canyon area was the closest ponderosa pine forest area having widespread unburned conditions. Samples were collected directly in 5-mL plastic vials for ^{137}Cs activity measurement. These samples were gathered to provide a range of likely concentration values of fallout ^{137}Cs in the decomposition layer of mature, unburned ponderosa pine forest areas.

The ^{137}Cs activity in study samples was measured using a high purity germanium γ ray detector system (EG&G ORTEC, Oak Ridge, TN). The detector uses a well configuration with a 76.4-mm-diameter crystal having a measured efficiency of 95.8% relative to a reference sodium iodide crystal. Sample count times averaged 26.8 h and counting error averaged 3.4%. The detector used 5-mL plastic vials that held on average 3.2 g of fractioned soil and sediment material.

RESULTS

Cesium-137 Increases on the Ground Surface after Fire

Average ^{137}Cs concentrations in the topmost 50 mm of post-fire soils on burned study plots (0.044 [$\pm$0.002; standard deviation] Bq g^{-1}) were about three times greater than in soils on unburned plots (0.015 [$\pm$0.003]

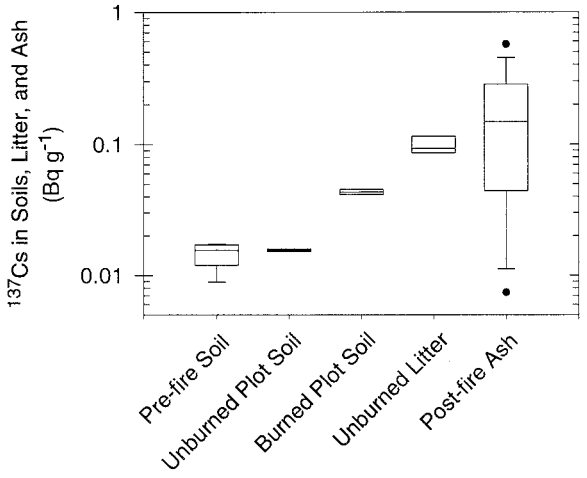

Fig. 2. Cesium-137 concentrations in the upper 50 mm of soil (before and after wildfire), unburned litter, and post-fire ash. Data modes are shown within the 25th to 75th percentile boxes. Whisker bars show standard deviation and outlier data are shown as points where present [ash data is from New Mexico Environment Department; pre-fire soils data from Los Alamos National Laboratory (2001)].

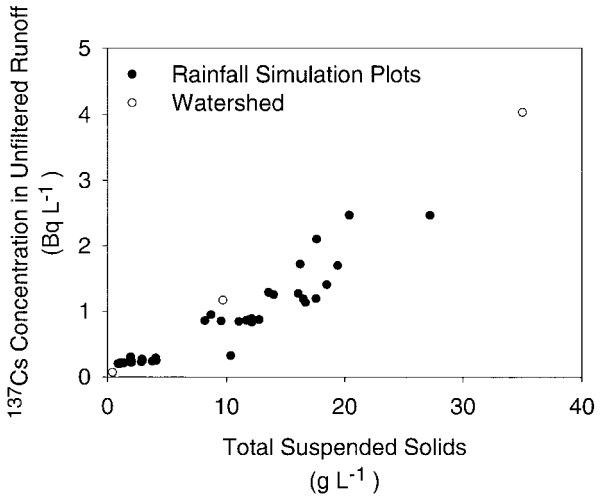

Fig. 3. Concentrations of ^{137}Cs and total suspended solids in runoff (r = 0.95).

Bq g^{-1}; Fig. 2). Similarly, the post-fire ^{137}Cs concentrations from burned plots were about three times higher than levels in the 1995–1999 pre-fire soil samples (0.014 [±0.003] Bq g^{-1}; Los Alamos National Laboratory, 2001). Concentrations within just the ash alone averaged 0.192 Bq g^{-1} with a range of 0.007 to 0.570 Bq g^{-1}. These ash concentrations were up to 40 times higher than concentrations in pre-fire soils (Fig. 2).

The ^{137}Cs concentrations in the litter organic decomposition layer from the closest widely unburned site at Los Alamos averaged 0.10 (±0.02) Bq g^{-1} (Fig. 2). Similarly, the ^{137}Cs concentrations in litter organic decomposition layer samples from three additional unburned ponderosa pine sites in New Mexico and Colorado averaged 0.09 (±0.07) Bq g^{-1}. By comparison, ^{137}Cs concentrations in laboratory-ashed samples of overstory and understory vegetation from the study site before the fire averaged about one order of magnitude less (0.014 [±0.020] Bq g^{-1} overstory, 0.010 [±0.016] Bq g^{-1} understory; Gonzales et al., 2000).

Cesium-137 Increases in Surface Water Runoff after Fire

Total yields of ^{137}Cs in runoff leaving plots during rainfall simulation (Bq transported per ha, per mm rainfall) were about one order of magnitude greater from burned plots (average for the three rain event simulations of 5.96 KBq ha^{-1} mm^{-1}) than yields from unburned plots (average of 0.55 KBq ha^{-1} mm^{-1}; Table 1). The total yields from individual burned plots ranged from 2.4 to 11.6 KBq ha^{-1} mm^{-1}. The highest yields occurred on plots having the least groundcover, with the greatest ^{137}Cs yield occurring on the plot with 80% bare soil compared with lower yields for plots with less bare soil (i.e., 69, 58, and 38%).

The post-fire concentrations of ^{137}Cs in runoff from burned plots averaged 1.22 (±0.44) Bq L^{-1}, while runoff from unburned plots averaged lower at 0.23 (±0.03) Bq L^{-1} (Table 1). Post-fire concentrations of ^{137}Cs in runoff from the Pajarito Watershed were elevated even higher, reaching 4.0 of Bq L^{-1}, an increase of more than two orders of magnitude over the pre-fire concentration average of 0.009 Bq L^{-1} measured between 1995 and 1999 (Los Alamos National Laboratory, 2001; Table 1). The ^{137}Cs concentrations in the unfiltered runoff samples were strongly correlated with amount of suspended solids present (r = 0.95) (Fig. 3). Total suspended solids measurements after the fire reached as high as 27 g L^{-1} for burned plots and 35 g L^{-1} in the runoff from the burned watershed, compared with an average of 2.30 g L^{-1} for unburned plots. Consistent with this, samples of unfiltered runoff from burned plots had much more ^{137}Cs sorbed to particulates. One-liter samples of runoff contained up to 2.3 Bq ^{137}Cs sorbed to particulates compared with a 0.16 Bq average in the separated water fraction.

Enrichment Effects

Enrichment of ^{137}Cs concentrations in runoff sediments occurred during surface water erosion on burned

Table 1. Average ^{137}Cs concentrations in runoff from July 2000 rainfall simulation.

Run	^{137}Cs in unfiltered runoff		^{137}Cs in sediment		^{137}Cs yield	
	Unburned	Burned	Unburned	Burned	Unburned	Burned
	Bq L^{-1}		Bq g^{-1}		KBq ha^{-1} mm^{-1}	
Dry	0.25 (±0.04)†	1.48 (±0.60)	0.032 (±0.012)	0.087 (±0.014)	0.55 (±0.09)	7.03 (±6.50)
Wet	0.22 (±0.01)	1.20 (±0.36)	0.034 (±0.005)	0.064 (±0.006)	0.41 (±0.01)	5.65 (±3.71)
Very wet	0.23 (±0.03)	1.03 (±0.19)	0.039 (±0.002)	0.060 (±0.004)	0.70 (±0.02)	5.20 (±3.45)
Average	0.23 (±0.03)	1.23 (±0.38)	0.035 (±0.006)	0.070 (±0.008)	0.55 (±0.04)	5.96 (±4.55)

† Values in parentheses represent standard deviations on multiple measurements.

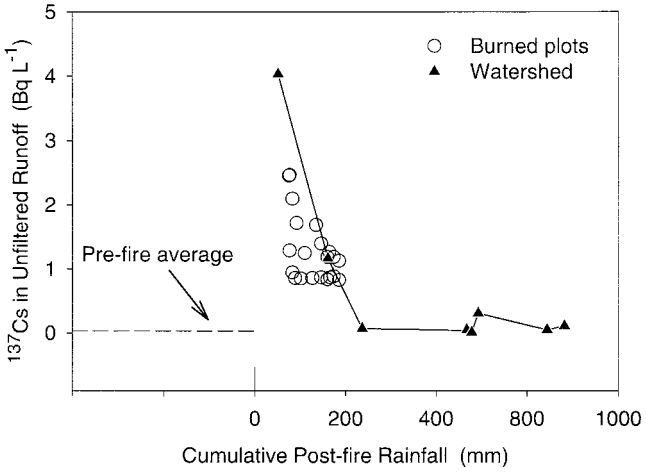

Fig. 4. Post-fire concentrations of ^{137}Cs in unfiltered runoff during cumulative rainfall.

and unburned study plots. Enrichment ratios comparing ^{137}Cs concentrations in runoff sediments to the concentrations in their parent soils ranged from 1.4 to 2.9. The average enrichment ratio for unburned plots was 2.3 (±0.6) and for burned plots was 1.6 (±0.4). The measured enrichment effect appears to be primarily associated with preferential entrainment of smaller-sized particles during surface water erosion. These smaller-sized particles (≤50 μm) made up greater weight percentages in runoff sediments than in parent soils (18.5% greater for unburned plots and 7.8% greater for burned plots). The smaller-sized particles also have greater affinity for the ^{137}Cs cation, resulting in higher per weight concentrations of ^{137}Cs in runoff sediments compared with parent soils (McHenry et al., 1973; Lane and Hakonson, 1982). This study did not distinguish between enrichment of organic and inorganic sediment material.

Duration of Elevated Cesium-137 in Runoff

In runoff from study plots, the highest ^{137}Cs concentration was found in the initial runoff, 2.46 of Bq L^{-1}, with subsequent concentrations lowered by half at the end of the rainfall simulations (Fig. 4). This decrease by half of the initial concentration occurred during application of about 120 mm of simulated rainfall, or about one-fourth of the average annual rainfall for the Los Alamos area. Similarly, for runoff from the upper portion of Pajarito Watershed, the highest concentration observed (4.0 Bq L^{-1}) was in a sample taken from the first substantial precipitation runoff event following the fire compared with lower concentrations in following events (Table 2). By the third watershed runoff event (23 Oct. 2000), 166 d following the fire, the measured concentration (0.07 Bq L^{-1}) was reduced to a level comparable with the pre-fire average (0.009 Bq L^{-1}).

DISCUSSION

Study data indicate increases in ^{137}Cs concentrations in the 0- to 50-mm of layer of soil following wildfire in ponderosa pine forest. The topmost layer of ash had the greatest ^{137}Cs concentrations compared with the entire 0- to 50-mm layer. This confirms with immediate post-fire measurements what Paliouris et al. (1995) suggested with years-later measurements—that burning of aboveground biomass during wildfires results in deposits at the ground surface having elevated ^{137}Cs levels.

At this study site, pre- and post-fire inventory measurements of ^{137}Cs were not made due to the unforeseen nature of the wildfire. However, concentrations in the post-wildfire ash averaged about one order of magnitude higher than that of ash from pre-fire samples of aboveground vegetation at the study site (Gonzales et al., 2000). This suggests that most of the ^{137}Cs in post-fire deposits did not come from burning of aboveground vegetation, but from burning of ground surface biomass (litter and decomposing litter). Measurement of ^{137}Cs in the litter decomposition layer at the nearest unburned ponderosa pine forest area indicated ^{137}Cs concentrations less than but close to the concentrations in post-fire ash (Fig. 2), further suggesting the ground surface organic matter as the primary contributor. At undisturbed, unburned ponderosa pine sites, the layer of decomposing litter is thought to provide a relatively stable reservoir of fallout radionuclides that when burned becomes the primary contributor to the elevated concentrations in residual ash. Confirmation of this will require thorough inventory measurements of pre- and post-wildfire conditions.

The concentrated ^{137}Cs in ash is susceptible to resuspension to the air by wind (Whicker et al., 2002) and subject to erosion and transport by water processes (Johansen et al., 2001b). This study reports ^{137}Cs concentrations in runoff from areas having burned conditions that were temporarily elevated more than two orders of magnitude greater compared with unburned areas. The ^{137}Cs was associated primarily with particulates in the runoff, with only a small fraction dissolved in the water fraction.

The occurrence of elevated fallout radionuclide levels following fire is expected to be relatively short lived. Study data indicate that the highest levels of ^{137}Cs appeared in initial runoff events, and measurements in

Table 2. Cesium-137 concentrations in runoff from the Pajarito Watershed.

Sampling date	^{137}Cs in unfiltered runoff†	Days from fire	Cumulative precipitation after fire
	Bq L^{-1}	d	mm
1995–1998, Pre-fire surface water and runoff samples	0.009 (±0.07)	pre-fire	NA
28 June 2000 (first runoff event)	4.03	48	51
9 Sept. 2000 (second event)	1.17 (±0.22)‡	121	160
23 Oct. 2000 (third event)	0.07 (±0.10)‡	166	236
February–August 2001	0.09 (±0.11)‡	264–454	467–882

† Los Alamos National Laboratory (2001). Values in parentheses represent standard deviations on multiple measurements.
‡ Counting uncertainty.

succeeding runoff indicated that elevated ^{137}Cs levels decreased rapidly. After approximately 240 mm of rainfall the ^{137}Cs concentrations approached unburned condition levels. When longer time frames are considered, this rapid redistribution of ^{137}Cs will appear as a sharp pulse of elevated concentrations leaving burned hillslope areas.

To place our results in a broader context, we compare data from rainfall simulation following wildfire in forest, with previous similar measurements in shrubland and grassland (Johansen et al., 2001a, b). The main similarities between these rainfall simulation studies were rainfall amount and intensity, rainfall duration, and size and shape of study plots. The main differences were vegetation types (grassland, shrubland, and forest), soil textures, and type of burning. An intense wildfire burned the ponderosa pine forest site, while controlled fire was used at the grassland and shrubland sites. Some variation existed in slopes (from 4.5 to 9%) of the various study plots (Johansen et al., 2001b). However, the steepest slopes (in grassland) did not produce the largest sediment yields or ^{137}Cs transport consistent with the expectation that these differences in gently sloped surfaces are unlikely to cause large differences in sediment yields (Wilcox et al., 1988).

Combined results show amplified ^{137}Cs transport following burning in ponderosa pine forest where the intense wildfire consumed up to 80% of the ground cover (Fig. 5). The ^{137}Cs transport was consistently lower from the unburned plots in all vegetation types and from the burned plots in grassland and shrubland where smaller percentages of ground cover were removed by fire. Further, the transport of ^{137}Cs relative to the percentage of bare soil for these plots is not linear, but appears to have little change as percent bare soil varies between 0% and about 60 to 70%, and sharp increases in ^{137}Cs transport when the amount of bare soil is greater than 60 to 70%. Correlation between ^{137}Cs yields and sediment yields was high across all sites ($r = 0.96$).

Part of the elevated levels of ^{137}Cs in runoff appears to be a result of the enrichment of ^{137}Cs in sediments during surface water erosion that can further concentrate the ^{137}Cs. Sediments leaving study plots had higher proportions of fine-grained particles than their parent plots, and corresponding higher amounts of ^{137}Cs. Enrichment by water erosion on plots in this study ranged from 1.4 to 2.9. This range is consistent with other enrichment ratio measurements of 0.4 to 7.1 made in studies that examined enrichment of sediments for a broad range of radionuclides and nutrients on varying landscape scales (Johansen et al., 2001b); and with measurements of 0.4 to 4.9 specific to ^{137}Cs in small watersheds (Weigand et al., 1998). Our study specifically examined the question of whether or not soil heating during fire would alter enrichment ratios in subsequent runoff. In repeated tests on burned and unburned plots in grassland, shrubland, and forest (this study and Johansen et al., 2001b), no significant difference in enrichment ratio was measured in sediments from burned and unburned parent plots ($p < 0.05$; Fig. 6). Enrichment occurred on both types of plots, and ^{137}Cs yields were much higher on burned plots because of the associated greater sediment and ash yields. However, the ratio of enrichment between runoff sediments and parent soils was about the same for burned and unburned surfaces.

The various concentration and redistribution mechanisms discussed above suggest that the temperature at

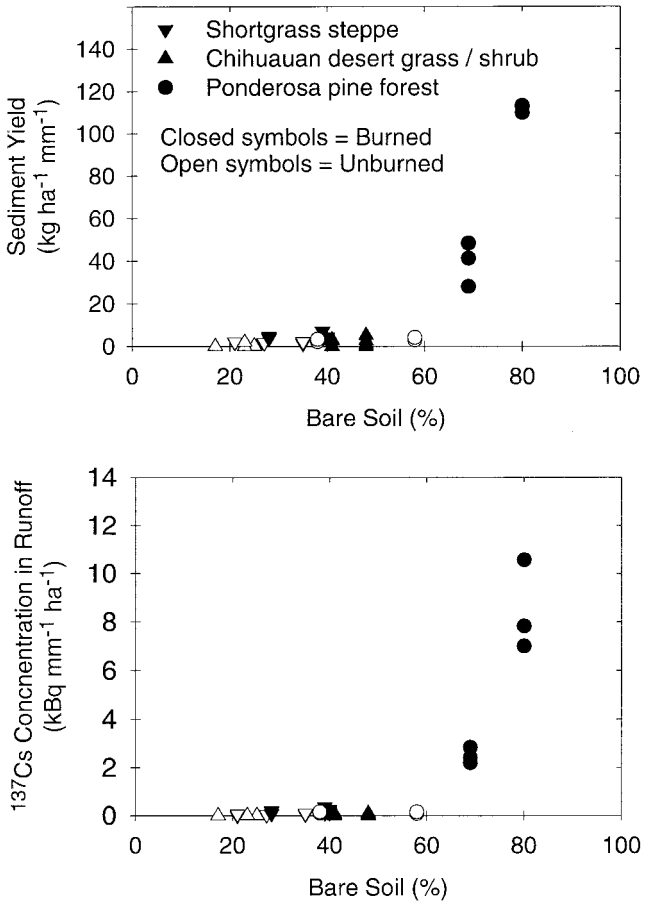

Fig. 5. Sediment yields and ^{137}Cs yields from rainfall simulations on burned and unburned plots over a range of vegetation types (sediment yield data from Johansen et al., 2001a,b).

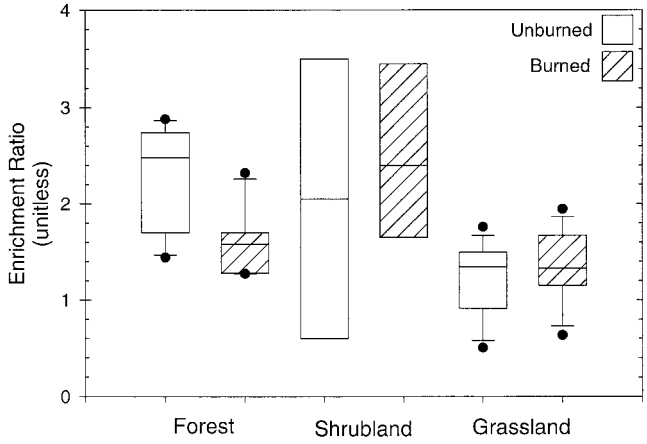

Fig. 6. Enrichment ratios in runoff sediments from rainfall simulation in a range of ecosystem types. Data modes are shown within the 25th to 75th percentile boxes. Whisker bars show standard deviation and outlier data are shown as points where present.

which fire burns may be a key underlying factor in determining the fate of post-fire ^{137}Cs. Higher fire temperatures are expected to lead to combustion of a greater portion of the ground surface litter layer that contains elevated concentrations of fallout radionuclides. The ^{137}Cs concentrations in ponderosa pine forest litter measured in this study (about 0.10 Bq g^{-1}) are about an order of magnitude greater than typical soil levels in the southwestern USA (about 0.01 Bq g^{-1}; Fresquez et al., 1996). In addition to ashing of ground surface litter where most fallout ^{137}Cs resides, higher combustion temperatures may remove greater percentages of ground cover contributing to accelerated redistribution by surface water runoff. Our results suggest that approximately 30 to 40% ground cover is needed to prevent highly accelerated erosion on slopes of less than 10%. Further research is needed to make conclusive statements relating fire temperatures and ^{137}Cs transport rates.

While this study focuses on concentration and transport of ^{137}Cs, our data provide insights for other particle-reactive fallout radionuclides, metals, and nutrients. Hulse et al. (1998) showed that ^{137}Cs behavior in soils was well correlated to that of ^{241}Am and ^{239,240}Pu. Furthermore, following the Cerro Grande fire, data gathered by the Los Alamos National Laboratory indicated order-of-magnitude concentration increases in ^{90}Sr, ^{238}Pu, ^{239,240}Pu, barium, manganese, and calcium (among others) in runoff samples collected in burned watersheds (Gallaher et al., 2002). Samples for these measurements were taken in watercourses upstream of the Los Alamos National Laboratory areas, away from laboratory waste areas.

CONCLUSIONS

This study documents short-term order-of-magnitude increases of fallout ^{137}Cs concentrations at the ground surface and in surface water runoff following a major fire in a southwestern U.S. ponderosa pine forest. The greatest transport of concentrated ^{137}Cs by surface water runoff occurred in the initial runoff event following fire and decreased relatively rapidly thereafter. These results indicate that fire concentrated ^{137}Cs in ash deposits, which was then redistributed away from burned areas during the rainfall events following fire. This redistribution can be viewed as a pulse of elevated concentrations of ^{137}Cs leaving the burned area when considering long time frames. Part of the concentration increases in runoff resulted from enrichment of ^{137}Cs during erosion, specifically the preferential entrainment of small particles having relatively high affinity for ^{137}Cs. Redistribution rates for ^{137}Cs increased sharply when fire removal of groundcover resulted in more than 60 to 70% bare soil on gently sloped plots. Study results provide new data on contaminant concentration and redistribution following wildfire in forests and systematic comparisons between forests, grasslands, and shrublands. This new information may be useful for a wide range of interests including ecosystem dynamics studies, geophysical studies, fire management, and risk assessments where periodic fire events may dominate contaminant redistribution rates.

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