

Organochlorine Compounds in a Brazilian Watershed with Sugarcane and Intense Sediment Redistribution

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ABSTRACT

Until 1985 persistent organochlorine compounds such as aldrin [(1 α ,4 α ,4a β ,5 α ,8 α ,8a β)-1,2,3,4,10,10-hexachloro-1,4,4a,5,8,8a-hexahydro-1,4:5,8-dimethanonaphthalene] and heptachlor (1,4,5,6,7,8,8-heptachloro-3a,4,7,7a-tetrahydro-4,7-methano-1H-indene) were recommended and used as insecticides on a large scale in sugarcane (*Saccharum officinarum* L.) cropping in Brazil. The environmental impact of these applications was not investigated in the past neither prior to nor after their restriction of use in Brazil. In a case study conducted during 1999, organochlorine insecticides were investigated in soils, colluviums, submerged sediments, and organisms in a watershed located in a traditional sugarcane-growing region in the southeastern region of Brazil. The results indicated that past applications of organochlorine insecticides (prior to 1985) do not represent an environmental threat at the present time. Most insecticides applied in the past were not detected or were present only in amounts that were below the detection limit. The organochlorine compounds lindane [(1 α ,2 α ,3 β ,4 α ,5 α ,6 β)-1,2,3,4,5,6-hexachlorocyclohexane] and heptachlor that remained on the market after 1985 for purposes other than for soil application were still detectable in significant amounts in soils, sediments, and soil organisms (heptachlor in form of the epoxide metabolites). This may suggest that these residues were originated from applications after 1985. These compounds showed a capacity for accumulation in sediments and soil organisms. The enrichment ratio from soils to sediments was 2 to 3 times and from soils to organisms up to 20 times.

THE world's sugarcane production area amounts to about 20×10^6 ha of land, of which 5×10^6 ha (25%) is located in Brazil. Most of these crops (65%) are concentrated in the southeastern region of Brazil (AGRIANUAL, 1999). Sugarcane is usually grown as a monoculture concentrated around the sugar and/or alcohol processing industries (and therefore, mostly close to urbanized areas). The application of insecticides for the control of termites and soil bug larvae is a routine practice. The insecticides are sprayed on the nursery stalks and surrounding soil in the planting furrow and/or placed as a continuous deep layer by positioning the application nozzles behind the plow (Novaretti et al., 1991). Organochlorine insecticides, mainly aldrin and heptachlor, were officially recommended and used until 1985

(IAA, 1982). After 1985, these products were restricted for soil application and substituted for about 3 yr by the chlorinated hydrocarbon endosulfan (6,7,8,9,10,10-hexachloro-1,5,5a,6,9,9a-hexahydro-6,9-methano-2,4,3-benzodioxathiepin 3-oxide) and more recently by the less persistent organophosphate terbufos (*S*-[[[(1,1-dimethylethyl)thio]methyl] *O,O*-diethyl phosphorodithioate) and the carbamate carbofuran (2,3-dihydro-2,2-dimethyl-7-benzofuranyl methylcarbamate). In the past, a lack of coordination and awareness among the government, industry, trade, extension, research, and pesticide consumers resulted in the absence of systematic research or monitoring of possible environmental or human-related impacts from the use of organochlorine substances on such large areas (Yorinori, 1983). Currently, most organochlorine research is kept on a large-scale basis, focusing on global distillation effects (Simonich and Hites, 1995), regional cold condensation processes (Blais et al., 1998), or highly contaminated areas (Haimi et al., 1992). The high environmental persistence (Edwards, 1976) of most organochlorine compounds was reported for tropical conditions (Grace et al., 1993). Significant amounts of heptachlor metabolites were still found 16 yr after application in soils of highly contaminated sites (Nash and Harris, 1973). In addition, the potential enrichment in sediments of surface-water resources (Burgoa and Wauchope, 1995; Ghadiri and Rose, 1993) makes it possible that "hot spots" or bioaccumulation processes may occur in Brazilian sugarcane-growing regions.

The objectives of this study were to (i) determine the remaining residues of organochlorine compounds in soil in a representative watershed of commercial sugarcane production, (ii) evaluate their redistribution in trapping positions such as floodplain colluviums and submerged sediments in a water reservoir, and (iii) estimate their current bioaccumulation potential. The selected study area was the Ceveiro watershed, located in a sugarcane-growing region in southeastern Brazil (Fig. 1).

MATERIALS AND METHODS

Study Area

The Ceveiro watershed (1990 ha) is located in the southeastern part of Brazil (Piracicaba) with central coordinates of 22°38'54"S and 47°45'40"W (Fig. 1). The climate, according to Koeppen's classification, is Cwa (i.e., humid subtropical with a dry winter and less than 30 mm rain in the driest month). The temperature in the hottest month is in excess of 22°C and in the coldest below 18°C. The landscape is usually composed of S-shaped profiles and the mean slope value is 13%. The soil types in the area are usually sandy at the surface and mainly represented by Ultisols, Alfisols and Entisols. The sandy surface and the steep slopes, together with intensive cropping, have resulted in high soil erosion rates (Weill et al., 1998) favorable for sediment and particle-adsorbed pollutant

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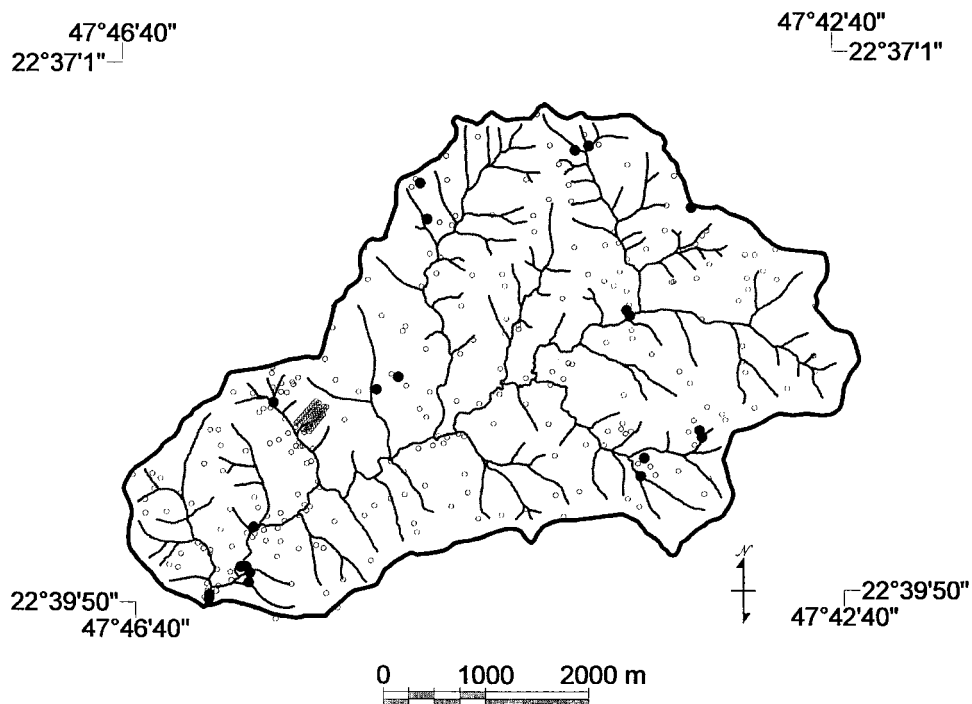


Fig. 1. Location of the samples used for previous soil erosion studies (outlined circles) and for the organochlorine analysis (filled circles).

redistribution. The floodplain positions are filled with recently deposited colluviums, characterized by a very initial successional stage of the vegetation, composed predominantly of grass. In recent years no significant land use changes have occurred, so the available data for land use for 1995 composed of sugarcane (66%), pastures (14%), forests (18%), and non-agricultural use (2%) represent the land use over the last 20 yr. Sugarcane is also the dominant crop in the region, covering 80% of arable land. At the lower part of the watershed, a water reservoir was constructed in 1982–1983 for providing fresh water for human consumption. The sediments trapped in this reservoir were originated from the time after the soil application of organochlorine pesticides was officially banned in 1985.

Interviews with local sugarcane producers and pesticide dealers indicated that in this watershed soil applications of pesticides have always followed the official recommendations. From the introduction of sugarcane in the area in the early 1960s, several organochlorine pesticides, mainly aldrin and heptachlor, were sprayed in the soil by sugarcane planting and renewal until 1985. The exact applied amounts, locations, and products could not be identified.

Soil, Colluvium, and Sediment Sampling

The samples used for this study are part of a larger set of samples. In total, 316 soil, colluvium, and sediment positions were collected and analyzed for chemical and physical properties important for soil erosion calculations (Sparovek et al., 2000). The objective of the selection of subsamples at 26 locations used for this study was to include the entire expected range of concentrations of organochlorine compounds in a smaller number of samples. The selection criteria were based on soil types (mainly surface soil clay content and soil organic matter), the calculated erosion rates, and the sedimentation conditions observed in the watershed. The trapping positions along the main stream (including colluvium positions very close to the sediment source, located in the middle of the area to the downstream submerged sediments) were also used as

criteria for location selection. The locations of the subsamples used for this study were highlighted among the total sampling locations and are shown in Fig. 1. This sampling procedure, based on the previous investigations about soil erosion and sedimentation processes (Sparovek et al., 2000), was designed to allow a comprehensive assessment of the influence of the erosion and sedimentation processes on the redistribution of organochlorine compounds with a relatively small number of samples. The submerged sediments were collected in two positions and in different depths of the reservoir, to make the study representative in relation to spatial variability and sedimentation period. One position (Sample 21) was located close to the inflow of the main stream into the reservoir and was sampled from 0 to 10 cm depth. The other position (Samples 22–26) was located close to the reservoir's outlet and was sampled at different depths, as indicated in Table 1. This procedure aimed to include a wide range of trapping positions (close to the main stream inflow position or the reservoir's outlet) and sedimentation periods (the deeper samples representative for sediments trapped close to the reservoir's construction period and the surface samples for more recent times) in the sediment subsamples. For statistical analysis the six sediment samples were considered as replications.

Soil (10 samples) and colluvium (10 samples) were collected during 1999 at a depth of 0 to 20 cm. The samples were stored frozen (-18°C) as collected from the field until they were prepared for analysis. The samples were dried at room temperature and sieved (2 mm) prior to analysis.

Sampling Organisms for Bioaccumulation Evaluation

The positions used for soil and colluvium sampling were checked for aboveground living organisms with the objective to obtain co-located samples of soils and colluvium and bioindicators from the soil's mesofauna. Organisms were sampled by digging and hand-sorting. Earthworms in sufficient numbers for analysis were found in seven locations and insect larvae in one. The earthworms were identified as *Pontoscolex corethrurus* (Müller, 1857) according to external anatomy,

Table 1. Organochlorine compound concentrations in soils, colluviums, sediments, and organisms from the Ceveiro watershed (Brazil).

Sample	Type	Depth	Lindane	Heptachlor-epoxide	
				cis	trans
		m	μg kg ⁻¹		
1	soil	0–0.2	4.0	<1	<1
	worm†	0–0.2	<2	2.0	<2
2	soil	0–0.2	<1	3.0	<1
	worm	0–0.2	<3	25.0	<3
3	soil	0–0.2	4.0	<1	<1
	worm	0–0.2	<3	<3	<3
4	soil	0–0.2	1.0	<1	<1
	worm	0–0.2	2.0	<2	<2
	larvae‡	0–0.2	23.0	41.0	3.0
5	soil	0–0.2	1.0	<1	<1
6	soil	0–0.2	1.0	<1	<1
7	soil	0–0.2	1.5	<1	<1
8	soil	0–0.2	2.8	<1	<1
9	soil	0–0.2	<1	<1	<1
10	soil	0–0.2	<1	1.7	<1
	̄ soil§		1.7a		
11	colluvium¶	0–0.2	4.0	<1	<1
	worm	0–0.2	<3	<3	<3
12	colluvium	0–0.2	<1	<1	<1
	worm	0–0.2	<3	<3	<3
13	colluvium	0–0.2	<1	1.0	<1
	worm	0–0.2	<3	<3	<3
14	colluvium	0–0.2	2.0	<1	<1
15	colluvium	0–0.2	1.4	<1	<1
16	colluvium	0–0.2	3.0	<1	<1
17	colluvium	0–0.2	3.0	<1	<1
18	colluvium	0–0.2	<1	<1	<1
19	colluvium	0–0.2	<1	<1	<1
20	colluvium	0–0.2	<1	<1	<1
	̄ colluvium		1.6a		
21	sediment#	0–0.1	2.0	1.0	<1
22	sediment	0–0.05	3.5	<1	<1
23	sediment	0.05–0.1	2.4	<1	<1
24	sediment	0.1–0.2	6.3	<1	<1
25	sediment	0.2–0.3	3.6	<1	<1
26	sediment	0.3–0.5	1.7	<1	<1
	̄ sediment		3.3b		

† Geophagous earthworm *Pontoscolex corethrurus* (Müller, 1857) expressed in dry matter (lyophilized).

‡ Bug larvae *Migdolus fryanus* (Westwood, 1863) expressed in dry matter (lyophilized).

§ Mean values calculated considering the measured rates or half the concentration of the detection limit. Different characters after mean values indicate significant (* or $p < 0.05$) differences according to Duncan test.

¶ Recently deposited sediments located at the floodplain positions along the main stream.

Submerged sediments deposited in a water reservoir at the watershed outlet position.

calm behavior after capture, and habitat, following the descriptions of Righi (1990). *Pontoscolex corethrurus* is the most abundant geophagous earthworm species in South America (Righi, 1990), migrates only a few meters each year (Gates, 1973), and is frequently observed in agroecosystems (Zou and Gonzalez, 1997), especially in sugarcane (Spain et al., 1990). Earthworms are accumulators of organochlorine compounds (Haimi et al., 1992), represent an important transfer mechanism of organochlorine compounds to the aboveground biota (Beyer and Gish, 1980), and are indicators in tests for chemicals and plant protection products in several countries (Kula, 1998). All these characteristics make *P. corethrurus* a valuable organism for indicating potential bioaccumulation of organochlorine substances in the study area. At one sampling point, larvae from *Migdolus fryanus* (Westwood, 1863) were found in sufficient quantities and collected for analysis. The organisms were stored frozen (–18°C) and lyophilized prior to the analysis.

Analytical Procedures

Soil (50 g), sediment (50 g), and organism (~1 to 3 g of lyophilized worm or larvae) samples were extracted with 200 mL of 1:1 cyclohexane–ethylacetate (Merck, Darmstadt, Germany) in a Soxhlet extractor for 16 h. The extracts were concentrated and filtered with a membrane filter (Macherey & Nagel, Düren, Germany) of 45 μm. The cleanup was performed according to the DFG S19 multimethod (DFG Pesticide Commission, 1987) by gel permeation chromatography (bio-beads S-X3 8 (Bio-Rad) cyclohexane–ethylacetate (1:1) and additionally by minisilica gel column chromatography, purified, cleaned up and eluted with toluene. The determination was carried out on a Hewlett–Packard (Palo Alto, CA) 5890II gas chromatography–electron capture detector with an injector temperature of 280°C and a detector temperature of 330°C. Analysis was repeated in DB-35 and DB-5 capillary columns (30-m length, 0.25-mm i.d., 0.25-μm film thickness; J&W Scientific, Folsom, CA) using helium as the carrier gas at 1 mL min⁻¹. Typical quantification limits for 50 g soil and sediments were 1 μg kg⁻¹ and for 13 g organisms ~2 to 3 μg kg⁻¹.

For quality assurance, fortification experiments were performed in duplicate for soil and worm samples. In these experiments, 50 g of soil was spiked at a concentration level of 1 μg kg⁻¹ and 1 g worm material at a concentration of 25 μg kg⁻¹. In soil samples the recovery rates for lindane, heptachlor, cis-heptachlorepoxyd, transheptachlorepoxyd, cischlordane, transchlordane (1,2,4,5,6,7,8,8-octachloro-2,3,3a,4,7,7a-hexahydro-4,7-methanol-1H-indene), and dieldrin [(1α,2β,2α,3β,6β,6α,7β,7α)-3,4,5,6,9,9-hexachloro-1a,2,2a,3,6,6a,7,7a-octahydro-2,7:3,6-dimethanonaphth[2,3-b]oxirene] were determined in the range of 70 to 110%. Each series of soil, sediment, or worm samples was accompanied by a blank sample.

A first screening test with 10 samples was analyzed and evaluated for HCB (hexachlorobenzene), lindane (alphaHCH, betaHCH, deltaHCH), PCNB (pentachloronitrobenzene), heptachlor, cisheptachlorepoxyd, transheptachlorepoxyd, alphachlordane, gammachlordane, oxychlordane, alphaendosulfan, betaendosulfan, endosulfansulfate, aldrin, dieldrin, endrin [(1α,2β,2αβ,3α,6α,6αβ,7β,7α)-3,4,5,6,9,9-hexachloro-1a,2,2a,3,6,6a,7,7a-octahydro-2,7:3,6-dimethanonaphth[2,3-b]oxirene], p,pDDE, o,pDDT, p,pDDD, p,pDDT, mirex [1,1a,2,2,3,3a,4,5,5,5a,5b,6-dodecachlorooctahydro-1,3,4-metheno-1H-cyclobuta[cd]pentalene], methoxychlor [1,1'-(2,2,2-trichloroethylidene)bis(4-methoxybenzene)] and tetradifon [1,2,4-trichloro-5-[(4-chlorophenyl)sulfonyl]benzene]. Other samples were only evaluated for the compounds present in levels above the quantification limit in the screening samples (the quantification limit was defined as five times the standard deviation of the background of the bulk sample).

RESULTS AND DISCUSSION

Although several organochlorine insecticides as dieldrin, cis- and transchlordane, oxychlorddane, lindane, and heptachlor metabolites could be qualitatively detected in soil, sediment, and worm samples, only heptachlor metabolites and lindane were present in amounts that were high enough for a reliable quantification (above quantification limit). The past intensive use of organochlorine pesticides and the narrow spectrum of compounds with low residues of 1 to 6 μg kg⁻¹ in soil, colluvium, and sediment samples (Table 1) indicate that degradation or removal processes reduced the contamination to a level comparable with that in areas where no intensive use was made. The semivolatile characteris-

tic of these compounds (Blais et al., 1998) associated with atmospheric redistribution (Simonich and Hites, 1995) spread the persistent organochlorine substances everywhere on the Earth. Low levels of a wide spectrum of organochlorine substances, as observed in the Cerveiro watershed, were also observed in other tropical regions where no intensive applications were known (Osibanjo et al., 1994). Based on this data we may conclude that, in this case study area, the past frequent use of organochlorine substances for sugarcane production does not represent a significant environmental threat at the present time. Nevertheless, a more detailed analysis of the compounds lindane and heptachlorepoxydes that fall in the quantification range raises some interesting observations.

In Brazil, the organochlorine pesticides were officially banned for soil application in 1985, and thus withdrawn from legal sale for this purpose. Currently, some organochlorine compounds are allowed for purposes other than for soil application and can therefore still be purchased. Heptachlor is allowed for the treatment of power and telephone pedestals or wire fence posts to prevent termite damage. Lindane is a traditional component of lotions, creams, and shampoos for the control of lice and mites in humans and in veterinarian products for ectoparasites. Other components, previously recommended for soil application, such as DDT, aldrin, and dieldrin, were completely restricted and withdrawn from commerce.

A wide range of organochlorine insecticides were used until 1985 in the area but only lindane and heptachlor metabolites could be detected in significant amounts. The persistence in soils for heptachlor is lower and for lindane is equivalent when compared with other formerly recommended compounds such as DDT, dieldrin, and aldrin (Edwards, 1976). The transfer mechanisms to trapping positions such as colluviums and sediments are similar for most organochlorine compounds (Burgoa and Wauchope, 1995). Lindane degrades quickly under anaerobic conditions and a half-life of around 30 d is reported for lindane in flooded soils (World Health Organization, 1991). The presence of lindane in the submerged sediments was significantly higher when compared with soils and colluviums (Table 1), with an enrichment ratio of ~2 to 3. For lindane and heptachlorepoxydes, bioaccumulation could be clearly observed in at least three samples of soil organisms (Samples 1, 2, and 4 from Table 1). In particular the bug larvae, with higher fat content as compared with the earthworms, showed accumulation factors of ~20 or more in relation to the surrounding soils.

Based on the interviews with the landowners and pesticide dealers during the investigations, we considered that the possibility that lindane and heptachlor were previously applied more intensively than others was improbable. It is more probable that these two products have been applied and redistributed after the restriction of the organochlorine soil applications. This may have occurred in at least two pathways. The first is that the remaining legal applications (i.e., veterinarian products and lice and mite lotions for lindane, or wood treatment for heptachlor) constitute a significant mech-

anism for dispersion into the environment. In this case the foreseen exclusively local action under current legal use would not occur and environmental dispersion should be considered also in this case. Another pathway could be the continuity of soil application, even if illegal. These products are still available on the market and their well-known high efficacy for soil pest control may encourage farmers to keep using them. The methods used in this case study do not allow the confirmation of one hypothesis or the other. Independent of which is the more important redistribution pathway, maintaining these compounds on the market is essential for the existence of both. The complete restriction of these compounds and substitution by products of similar efficiency but lower bioaccumulation or danger for living organisms would probably bring detection levels to zero in a few years, as has occurred with other banned organochlorine compounds.

CONCLUSIONS

Past applications of organochlorine insecticides (prior to 1985) do not represent an environmental threat at the present time in the case study area located in a traditional sugarcane growing area in southeastern Brazil. Organochlorine compounds still available in commerce after 1985 for purposes other than soil application were detectable in significant amounts in soils, sediments, and living organisms showing a high capacity of bioaccumulation. A complete marketing ban is probably the best solution for avoiding the dispersion of these products into the environment.

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Hydrologic Response and Radionuclide Transport Following Fire at Semiarid Sites

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ABSTRACT

Infrequent, high-impact events such as wildfires, droughts, biological shifts, floods, and mechanical disturbances can greatly change land surfaces, including vegetative cover and soil characteristics, which in turn can trigger high rates of hydrologic erosion and associated transport of sediments and sediment-sorbed contaminants. Where persistent soil contamination exists, infrequent mobilization of contaminants may dominate in determining long-term risks to human and ecological receptors. Among these infrequent events, fire stands out as having the capacity to cause large increases in sediment transport. This study measured runoff, sediment yield, and mobility of sediment-sorbed contamination (¹³⁷Cs) on burned and unburned plots at the Waste Isolation Pilot Plant, New Mexico (WIPP), and the Rocky Flats Environmental Technology Site, Colorado (RFETS). Results showed that ¹³⁷Cs transport from burned plots was up to 22 times greater than that from unburned plots at WIPP and 4 times greater at RFETS. Associated runoff was up to 12 times greater on burned plots at WIPP and sediment yields up to 6 times greater. Further, ¹³⁷Cs concentrations in transported sediments were enriched compared with parent soils (expressed as enrichment ratio) by a factor of 2.3 at WIPP, and 1.3 at RFETS. However, enrichment ratios were not significantly different in sediments from burned and unburned plots. Our results provide new data on the effects of fire on the transport of sediment-sorbed contaminants, and demonstrate that rare events such as fire can greatly increase contaminant mobility.

RISK levels posed by contaminants in the environment are largely determined by basic processes such as erosion and sediment transport that can mobilize contaminants from source areas and carry them to receptors. These erosion and contaminant transport processes vary

over time and may be subject to large shifts induced by infrequent, high-impact events such as wildfire, drought, biological activity, mechanical disturbances, floods, and extraordinary wind or precipitation events. However, such events are not routinely incorporated into contaminant risk assessment, or only cursorily so, mainly due to lack of information on how they affect basic contaminant mobility. Further, infrequent disturbances that induce episodes of accelerated contaminant movement are not included in most risk assessment models that typically assume steady-state surface conditions and thus may underpredict risk over long time frames (Whicker et al., 1999).

Specific examples of infrequent events that triggered concerns about increased contaminant mobility occurred in the summer of 2000 when forest and rangeland wildfires burned in the western USA at three nuclear weapons facilities. Wildfires occurred at the Los Alamos National Laboratory, New Mexico (approximately 3000 ha burned), the Hanford Site, Washington (approximately 24 300 ha burned), and the Idaho National Engineering and Environmental Laboratory, Idaho (approximately 16 000 ha burned). These wildfires burned on and near radiological waste areas and raised heightened concerns over post-fire transport of radiological contamination by wind and water. As a result, new risk assessments were initiated to assess if accelerated transport of contaminants was occurring in post-fire conditions, and if risk levels were raised by fire. Further, these risk assessments are being conducted in the context of an increasingly accepted view that at nuclear weapons sites, radiological and nonradiological wastes will remain, posing potential risk to humans and the environment for tens or even hundreds of thousands of years (National Research Council, 2000). Over these long time frames, wildfire can reoccur many times in semiarid landscapes (Swet-

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