

WM2009 Conference, March 1-5, 2009, Phoenix, AZ

PLANT MOUNDS AS CONCENTRATION AND STABILIZATION AGENTS FOR ACTINIDE SOIL CONTAMINANTS-9175

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ABSTRACT

Plant mounds or blow-sand mounds are accumulations of soil particles and plant debris around the base of shrubs and are common features in deserts in the southwestern United States. An important factor in their formation is that shrubs create surface roughness that causes wind-suspended particles to be deposited and resist further suspension. Shrub mounds occur in some plant communities on the Nevada Test Site, the Nevada Test and Training Range (NTTR), and Tonopah Test Range (TTR), including areas of surface soil contamination from past nuclear testing.

In the 1970s as part of early studies to understand properties of actinides in the environment, the Nevada Applied Ecology Group (NAEG) examined the accumulation of isotopes of Pu, ²⁴¹Am, and U in plant mounds at safety experiment and storage-transportation test sites of nuclear devices. Although aerial concentrations of these contaminants were highest in the intershrub or desert pavement areas, the concentration in mounds were higher than in equal volumes of intershrub or desert pavement soil. The NAEG studies found the ratio of contaminant concentration of actinides in soil to be greater (1.6 to 2.0) in shrub mounds than in the surrounding areas of desert pavement. At Project 57 on the NTTR, 17 percent of the area was covered in mounds while at Clean Slate III on the TTR, 32 percent of the area was covered in mounds. If equivalent volumes of contaminated soil were compared between mounds and desert pavement areas at these sites, then the former might contain as much as 34 and 62 percent of the contaminant inventory, respectively. Not accounting for radionuclides associated with shrub mounds would cause the inventory of contaminants and potential exposure to be underestimated. In addition, preservation of shrub mounds could be important part of long-term stewardship if these sites are closed by fencing and posting with administrative controls.

INTRODUCTION

An alternative being evaluated by the United States (U.S.) Department of Energy (DOE), National Nuclear Security Administration Nevada Site Office (NSO/NNSA) for regulatory closure of areas of surface soil contamination created by past nuclear testing is fencing and posting with administrative controls. The Environmental Restoration Project, Soils Sub-Project “Corrective Action Units (CAUs)” occur primarily on the Nevada Test Site (NTS), the main continental location where the U.S. conducted nuclear testing, 105 kilometers northwest of Las Vegas, Nevada. However, some CAUs also exist on the Nevada Test and Training Range (NTTR) which surrounds a large portion of the NTS, including a portion of the NTTR managed as the Tonopah Test Range (TTR).

One category of Soil Sub-Project CAUs dominated by isotopes of Plutonium (Pu), Uranium (U), and Americium-241 (^{241}Am) is the “Pu dispersal experiment sites” [1]. These tests include “safety experiment sites,” where tests were conducted to determine if a weapon or warhead damaged in an accident would detonate with a nuclear yield, even if some or all of the high explosive components burned or detonated. Other Pu dispersal experiments evaluated whether nuclear devices could go critical during a transportation accident or if damaged during storage. These tests demonstrated that without all of the high explosives detonating simultaneously, criticality would not occur. However, these tests did result in widespread areas of near-surface soil contamination dominated by Pu, U, and ^{241}Am . For example, Clean Slate III on the TTR covers over 1,200 hectares above approximately 1.5 Becquerels per gram of soil (Fig. 1). Because the actinides were not consumed during Pu dispersal experiments, they were not incorporated into larger-size, “trinity glass” particles as occur at many other atmospheric test sites where criticality was achieved. The concern with the Pu dispersal experiment sites is primarily their inhalation risk because a significant fraction of the Pu particles (up to 40 percent) can be in respirable size fractions (less than 15 microns aerodynamic diameter) [2].

In addition, at most Pu dispersal experiment sites, a high percentage of the Pu activity as well as that of ^{241}Am and U is associated with accumulations of soil and plant debris around the base of plants, most commonly large, perennial shrubs [3]. Varyingly called “shrub mounds,” “blow sand mounds,” or “fertile islands,” these natural features are common in arid and semi-arid regions in the western U.S. (Fig. 2). In addition, they are also a major factor in stabilizing and preventing suspension of contaminants at Pu dispersal experiment sites [3,4]. At contaminated soil sites that have been mechanically disturbed, resuspension at Pu dispersal experiment sites can be three to four orders-of-magnitude higher than at soil sites associated with atmospheric tests where criticality occurred [5]. The concentration of actinides in the shrub mounds at particular Soil Sub-Project CAUs on the NTS, TTR, and NTTR has important implications for their characterization and post-closure management if they are closed by fencing and posting with administrative controls.

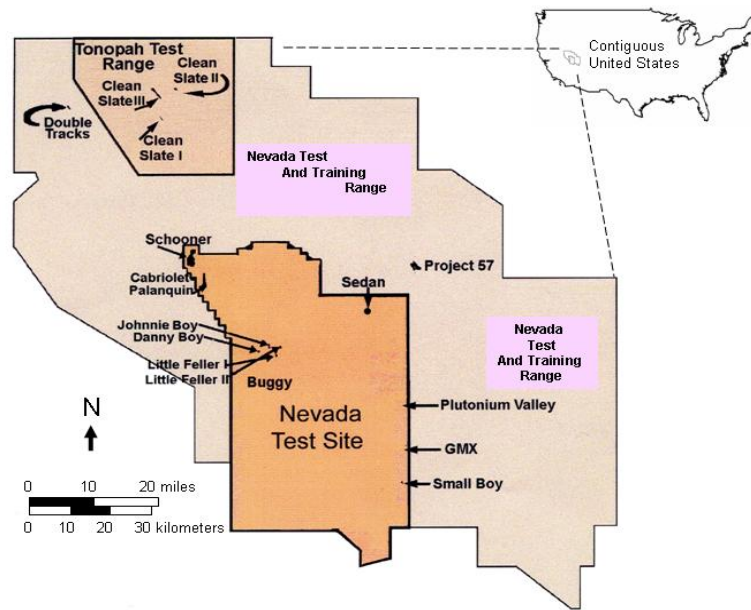


Fig. 1. Location of some of the major Soil Corrective Actions Units on the Nevada Test Site, Tonopah Test Range, and Nevada Test and Training Range.

FORMATION AND PROPERTIES OF SHRUB MOUNDS

Although shrub mounds occur in desert regions throughout western North America, they are more common in the Great Basin and Great Basin-Mojave Desert transitional zone region of the NTS, TTR, and NTTR. Several processes contribute to their development. Crawley and Nickling [6] cite the importance of shrubs creating surface roughness, which reduces the wind shear across an area, causing particles in suspension to be deposited. For clay- and silt-size particles in the mounds, the shrubs help to deposit particles whose origin are often relatively distant sources such as playas [7]. For sand-size particles and high-density radionuclide particles such as Pu oxides, the primary mechanism of migration and entrapment around the base of shrubs is probably by “saltation,” a process where larger particles “bounce” in low trajectories near the surface [8] (Fig. 3). The importance of saltation for the distribution and redistribution of radionuclide particles (and the role of plants in eventually stabilizing them) was demonstrated by tracking their migration after the Schooner cratering test on the NTS in 1969 [5]. At Schooner, periodic redistribution of radionuclides deposited near the test site continued after the

detonation for more than 30 days. Much of the migration was attributed to a combination of saltation and soil creep in areas where vegetation cover existed that acted to shelter particles from distribution between higher-velocity wind events. Essington *et al.* [3] found shrub mounds covered between 15 and 30 percent of the area. In addition, mound types were identified based on the vegetation around which they developed. Major



Fig. 2. Bioturbated shrub mound formed around a creosote bush (*Larrea tridentata*) on Yucca Flat on the Nevada Test Site. The photo was taken in June 2008.

categories included grass mounds, shrub mounds, and “complex mounds,” the last category being large mounds that formed around a group of shrubs of different species. The mounds vary in size. Gomes [9] measured shrub mounds as large as 3 meters (m) across and 0.30 m high in Plutonium Valley on the NTS. Those on the TTR are smaller, the largest being 2 m across and 0.20 m high, the smaller ones 0.3 m across and 0.05 m high [3].

In addition to being geomorphic features, shrubs mounds are often focal points of biological activity, particularly by small mammals, the origin of the “fertile island” terminology among ecologists. Compared to “intershrub” spaces, plant mounds, including those associated with many of the Pu dispersal experiments sites, are heavily bioturbated. Factors that favor small mammals living within shrub mounds are the mixture of particle grain sizes, making them easier to burrow into; the shade offered by the shrubs, seeds and leaf-litter in the mound that are a food source; and greater cycling

and concentrations of soil nutrients, particularly nitrogen and phosphorus [10,11]. In turn, bioturbation provides a positive feedback to the plants by allowing easier infiltration

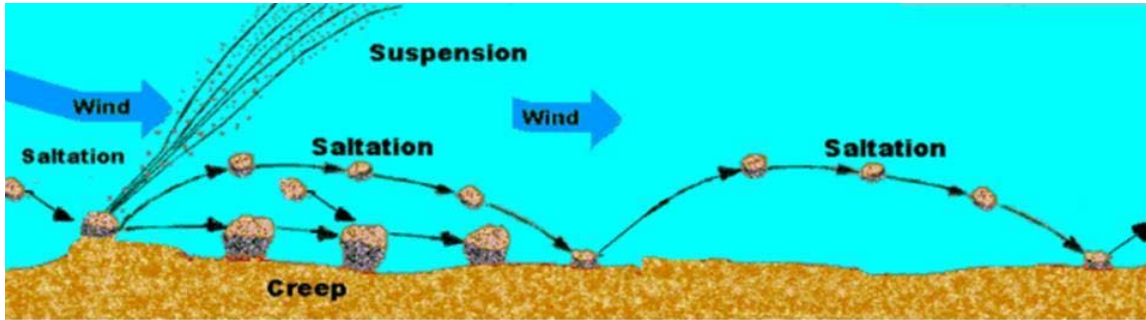


Fig. 3. Schematic of different types of particle suspension, including saltation, a process suspected to account for migration of higher density actinide particulates.

of water to the root zone of the plants when precipitation occurs. Shafer *et al.* [12] found saturated hydraulic conductivity (K_s) associated with plant mounds on the NTS to be five-fold greater than in intershrub areas. Other studies in the southwest U.S. have found even greater differences, such as Young *et al.* [13], who found K_s under the canopy of shrubs to be 100 times greater than in adjacent intershrub areas in the Providence Mountains in the Mojave National Preserve in California.

SHRUB MOUNDS AND ACTINIDE DISTRIBUTION IN SOIL

The Nevada Applied Ecology Group (NAEG), as part of its studies of factors affecting radionuclide migration and biological uptake properties at different types of nuclear tests on the NTS, NTTR, and TTR, first identified the significance of shrub mounds for actinide stabilization and distribution, particular at Pu dispersal experiment sites. The NAEG was formed in 1970 as an outgrowth of the formation of the Office of Effects Evaluation of what was then the DOE Nevada Operations Office. The objectives of the NAEG programs were to: (1) delineate locations of contamination; (2) determine concentrations in ecosystem components; (3) quantify rates of movement among ecosystem components; and (4) evaluate potential dose from Pu isotopes and other radionuclides [5,14].

Essington *et al.* [3] provided the most detailed comparison of radionuclide contaminants in shrub mounds versus the intershrub, desert pavement-covered spaces with studies at the Clean Slate III Soil Subproject CAU on the TTR, and Project 57 on the NTTR. They compared the concentration of total amounts of $^{239+240}\text{Pu}$, ^{241}Am , and total U in the top 5 centimeters (cm) of shrub mounds (“mound top”), in soil to a depth of 5 cm below the land surface datum beneath shrub mounds (the “mound bottoms”), and in adjacent intershrub spaces or “desert pavement” areas to the same depth. Probably because of bioturbation and mechanical mixing by wind, the concentration of per unit volume of the majority of plant mounds at Clean Slate III and Project 57 were found to be fairly uniform throughout their profile. In contrast, up to 85 percent of $^{239+240}\text{Pu}$ and ^{241}Am was concentrated in the top 2.5 cm in the intershrub or desert pavement areas of Pu dispersal

experiment sites [2,3]. This percentage increased to as high as 99 percent when the top 5 cm of soil were included. When measuring areal concentration, the mound bottoms and desert pavement areas had higher activity levels than the plant mounds. However, the concentration of radionuclides in plant mounds was higher in part because mixing of the top 2.5 cm of soil with soil at the depth interval of 2.5 to 5.0 cm diluted soil concentration in desert pavement areas.

Essington *et al.* [3] estimated that shrub mounds covered 31 percent of the radiologically Contaminated Area at Clean Slate III, and 17 percent at Project 57. At Clean Slate III, when the plant mounds “tops” were examined, it was determined that 29 percent of the inventory of $^{239+240}\text{Pu}$, ^{241}Am and total U was associated with shrub mounds, a ratio of inventory to area of 0.91. Similarly, at Project 57, 15 percent of the radionuclide inventory was associated with mound tops, a ratio of inventory to area of 0.85. When the concentration of $^{239+240}\text{Pu}$ and ^{241}Am in mounds and desert pavement areas were compared, ratios for three types of mounds (grass, shrub, and “complex”) ranged from 1.9 to 2.3 (average 2.0) at the 95 percent confidence interval with linear correlation R values between 0.82 and 0.93 (average 0.88). If equivalent volumes of soil were compared between mounds and desert pavement areas, then the former might contain as much as 62 percent of the inventory at Clean Slate II, and 34 percent at Project 57.

Brady [15] made similar studies of radionuclide contamination in vertical profiles of plant mounds and intershrub areas in Plutonium Valley on the NTS. At this site, the concentration of ^{241}Am in the top 5 cm of the intershrub or desert pavement areas was higher than the top 5 cm of the shrub mounds. However, when the total vertical profile of the shrub mound was compared to a comparable profile of intershrub area soil, then the total inventory ratio of ^{241}Am in the mound to the intershrub area was 1.6.

In general, these historic studies indicated that considerable concentration of $^{239+240}\text{Pu}$, ^{241}Am , and total U had occurred in shrub mounds at most of the Pu dispersal experiment sites. Not accounting for the contaminants in and beneath plant mounds would result in an underestimation of the inventory and potential exposure at these CAUs. In addition, the role of shrub mounds in stabilization of actinides at these sites is an important factor in long-term stewardship of these sites if they are closed in place.

STABILITY OF SHRUB MOUNDS

Although the shrub mounds act to prevent wind suspension of the radionuclide particles and other material within them, it is not uncommon to find mounds without supporting plants. Plant dieback probably occurs most frequently because of drought conditions. However, mounds where plant dieback has occurred can remain stable. One factor that may contribute to this is the high calcium carbonate (CaCO_3) content of silt and clay size particles that have and are being deposited and incorporated into shrub mounds today [7]. The CaCO_3 may act as a cementing agent, helping to form a thin crust over the surface, limiting erosion of the shrub mounds even if the plants around which they formed are temporarily lost. Also important in their role as long-term agents of radionuclide stabilization is that during wet cycles in arid regions, the shrub mounds acts as plant

recruitment or regrowth sites [16,17]. Over a 12-year period, Beatley [18] measured 14 percent death and 20 to 30 percent recruitment by seedlings or vegetation propagation from roots because of fluctuations in the timing and total precipitation in the northern Mojave-Great Basin transition region in Nevada. In addition, plants recruited to shrub mound sites following diebacks tended to be the same species around which the mound had originally developed. Because plant mounds serve as faster recruitment sites for new shrubs or regrowth of shrubs from rootstocks, revegetation may occur sooner on shrub mounds (at contaminated areas and otherwise) when an area is disturbed. This factor is of significance now because of the ongoing drought in the southwest U.S. where, with the exception of one year, total annual precipitation has been below normal since 1999.

CHANGING DISTURBANCE REGIMES FOR SOIL SITES

A disturbance factor that may make plant mounds (and concomitantly, actinides associated with them) less stable is an increase in wildfire. In some mid-elevation (above 800 m) parts of the northern Mojave Desert, an historic return interval of approximately 100 years for range fires may be in the process of replacing one where fires reoccur as frequently as every 20 to 30 years [19]. In the Great Basin, the return intervals appear to be even shorter. Several contributing factors are recognized. A sharp increase in fire frequency and size throughout the western U.S. beginning in the mid-1980s appears to correlate with an increase in average spring and summer temperatures [20]. This may be contributing to earlier loss of soil moisture and longer periods of dry plant biomass (particularly from annual plants) on the surface. In the western U.S., Westerling *et al.* [20] determined average spring and summer temperatures for the period 1987 to 2003 to be 0.87 °C higher than the period 1970 to 1986. Another factor contributing to greater fire frequency across the Great Basin and Mojave deserts has been the spread of introduced plants, most significantly cheatgrass (*Bromus tectorum*) and red brome (*Bromus rubens*) [21]. Today, these grasses can rapidly invade an area after it is disturbed such as by a fire. In many plant communities, they colonized interspaces between shrubs, increasing the total fuel load and allowing fires to move more easily between shrubs [19]. Both grasses were found locally on disturbed sites as early as the 1960s on the NTS, and they spread across the region of the NTS, NTTR, and TTR during a period of above-average precipitation in the 1980s [21]. Even in otherwise undisturbed sites, Shafer *et al.* [12] found individual *Bromus* sp. plants having become established on shrub mounds in Yucca Flat on the NTS because of the natural disturbance by bioturbation.

The only major Soil Sub-Project CAU to have burned over, the Palanquin Site, did so in 1977 [22]. Today, although some native grasses are present, there is a dense cover of *Bromus* sp. within intershrub areas of the radiologically Contaminated Area at Palanquin. Given the long half life of some of the radiological contaminants (e.g., 24,100 years for ²³⁹Pu), and factors increasing the frequency of fires, other Soil Sub-Project CAUs, including Pu dispersal experiment sites, may burn as well. Investigations on the impact of fires on the stability of contaminated soil sites, including Pu dispersal experiment sites, is underway as part of the NNSA/NSO Soils Sub-Project.

IMPLICATIONS FOR ENVIRONMENTAL MANAGEMENT OF SOIL CAUS

For the first of the Soils Sub-Project CAU where a fencing and posting closure alternative is being considered, the NNSA/NSO has proposed to the State of Nevada a risk-based closure standard of 25 millirems per year (mrem/yr) Total Effective Dose Equivalent for a site worker as demarcation criteria for the site boundary. Based on site characterization to date at atmospheric test sites where criticality occurred, potential dose contributing to the 25 mrem/yr standard will probably be dominated by direct exposure from fission products.

However, at Pu dispersal experiment sites, inhalation and ingestion may be greater contributors to potential exposure because of higher concentrations of actinides. Most of the data presented in this paper are from historic studies. It would be warranted to selectively sample some plant mounds and adjacent intershrub spaces to ascertain that the pattern of distribution of radiological contaminants as described in NAEG studies is still accurate. If soil sampling is part of the strategy for characterizing these sites, samples would need to be collected from shrub mounds as well as intershrub areas so as to not underrepresent contaminants of concern. At the Pu dispersal experiment sites, supplementing soil samples with measurement techniques that integrate activity levels over larger areas such as *in situ* spectral gamma, in which ^{241}Am would serve as an indicator for Pu isotope distribution may be a method of estimating potential inhalation and ingestion exposure [23].

In post-closure management of the Pu dispersal experiment sites, protecting shrub mounds could be an important component of long-term stewardship for these and potential other Soil Sub-Project CAUs. For example, in 2005 and 2006, organic emulsions were tested as to their effectiveness in preventing wind suspension of $^{239+240}\text{Pu}$ at the Smoky Site on the NTS [24]. However, if such agents are used, it would be important when applying them that the natural soil crusts that can form on plant mounds are not disturbed. The same precautions would be warranted if heavy machinery was used as part of reseeding a contaminated site. An alternative to treating an entire area with a soil stabilization agent would be to focus on areas of higher radionuclide concentration, including individual plant mounds. Maintaining native vegetation around Pu dispersal experiment CAUs could be important as well. Vegetation adjacent to the CAU may trap migrating Pu particles should the CAU be disturbed, particularly if the primary resuspension mode is by saltation. This would limit an increase in areal extent of contamination that could occur after a disturbance event.

ACKNOWLEDGEMENTS

Much of the information presented herein was developed as part of the Soils Sub-Project of the Environmental Restoration Project at NNSA/NSO. Jeffrey Tappen assisted with review of NAEG data. The work was performed by the Desert Research Institute (DRI) of the Nevada System of Higher Education under contract number DE-AC52-06NA26383 for the U.S. DOE.

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