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Viability of Mercury Phytoremediation in the former Terlingua Mining District, Big Bend, Texas, U.S.A.

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Introduction

The impacts of abandoned mercury (Hg) mine waste are a global concern. Currently there no active mercury mines in the United States (Gray 2006). The demand has diminished for mercury due to its toxicity and considerable recycling efforts (Sznopek and Goonan, 2000). In Terlingua, extraction of Hg was heated in a retort furnace to a temperature between 600 – 700 ^{0}C, which transforms the Cinnebar (HgS) to elemental Hg (Hg^{0}), which is the commercial grade Hg (Bailey and Phoenix, 1944). The retort process is inefficient, and the result is mine waste found around the mines contain non-converted cinnabar. Estimates indicate as much as a 25% loss of Hg during processing which has led to elevated concentrations of Hg in the surrounding environment (Gosar et all., 1997). Many of the Hg compounds are water soluble such as chlorides, oxychlorides, and sulfates. These contaminates have the potential to be released into the environment in mine runoff (Gray et al., 2004).

The former Terlingua Mining District in the Big Bend of Texas was an area of extensive mercury (Hg) mining between 1886 and 1973 (Burcham and Smith, 2013). At one time The Mining District was an area consisting of over 30 separate mines (Ross, 1941). The Chisos Mine was the largest and longest running mine in The Big Bend area. The mine was in operation from 1903 until 1943 where it closed due to the lack of cinnabar ore from which mercury is extracted (Burcham and Smith, 2013). To this day, the tailings or the waste rock left over from separating the economically valuable mercury from the gangue or worthless rock material is visibly present. These tailings piles have been left to the elements approaching 130 years. Mercury is considered toxic to all biota (Cherry and Guthrie, 2007) (Moreno-Jime´nez et al., 2005).

These mercury tailings are distributed into the surrounding environment by both strong winds and heavy seasonal rains (Mendez and Maier, 2008). The eolian process disperses the contaminated soil throughout the environment due to the lack of vegetation. The aqueous erosional processes carry the mercury contaminated soil from the tailings piles to the ephemeral creeks, to the perennial creeks, to the Rio Grande River, and eventually into the Gulf of Mexico.

The Lower Rio Grande Valley or The Valley of Texas is a name given to the lower portion of the Rio Grande River before it empties into the Gulf of Mexico (Vigness and Odintz, 2013). The Valley is an extensive farming area consisting of over 40 types of food crops as well as major producer of cotton (Texas A&M Agrilife). Water from the Rio Grande River is used to irrigate these crops. At the mouth of the Rio Grande, where the river empties into the Gulf of Mexico, there are a number of aquaculture farms. These farms raise freshwater fishes, saltwater fishes, and shrimp (Texas Aquaculture Association). The aquaculture farms are directly or indirectly using water from the Rio Grande River. Not to mention, the drinking water used by humans and livestock from the Rio Grande. The population using The Rio Grande River as their main source of drinking water from Big Bend National Park, TX to Harlingen, TX is roughly 1.6 million people on the U.S. side of the border (U.S. Census Bureau). The highest of environmental concerns associated with abandoned Hg mines are the downstream transportation of Hg. The most toxic of the Hg compounds are the chemical and microbial Hg metamorphosis into water soluble methyl-Hg.

The water runoff from the contaminated spoils piles does affect both terrestrial and aquatic life. Mercury bioaccumulates through the food chain, as predators eat other animals; the predator then absorbs the mercury of the prey, the mercury accumulates in muscle and organs of the predator (Government of Canada)(Gray, 2003). For humans, fish contamination is the highest percentage of mercury intake and poisoning (Fig. 1).

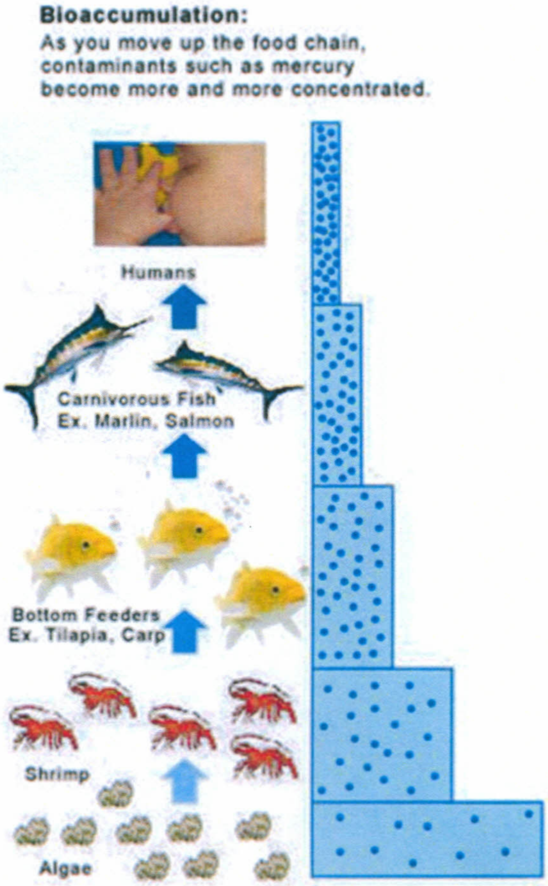

Figure 1: Bioaccumulation of mercury up the food chain. http://withfriendship.com/images/i/40337/

Mercury poisoning causes an array of health affects which some are irreversible even after exposure has ceased (Wheeler, 1996). Symptoms of the toxicity of mercury are tremors, ataxia, paresthesia, sensory disturbances, cardiovascular collapse, severe gastrointestinal damage, irreversible damage to the brain, kidneys, and developing fetuses, and even death (Raskin and Ensley, 2000) (Henry, 2000). Cell mutation, neurological, and kidney damage all can be passed on to offspring (Crinnion, 2000). In more recent findings, low-dose long-term exposure to Hg has been linked to

Alzheimer's and dementia (Hock et al, 1998)(Shcherbatykh and Carpenter, 2007)(Bocca et al., 2006), (Gerhardsson, 2008)(Gerhardsson et al., 2008). Therefore, the remediation of mercury from the former Terlingua Mining District should be proposed.

Background

Terlingua and The Big Bend Area of Texas reside in the northwestern portion of the Chihuahuan Desert (Fig. 2) with an average yearly precipitation rate of approximately 15.34 inches (National Park Service, 2013). This area is considered a Mid-latitude semi-dry or semi-arid region by the Koeppen climate classification system (Aguado and Burt, 2013).

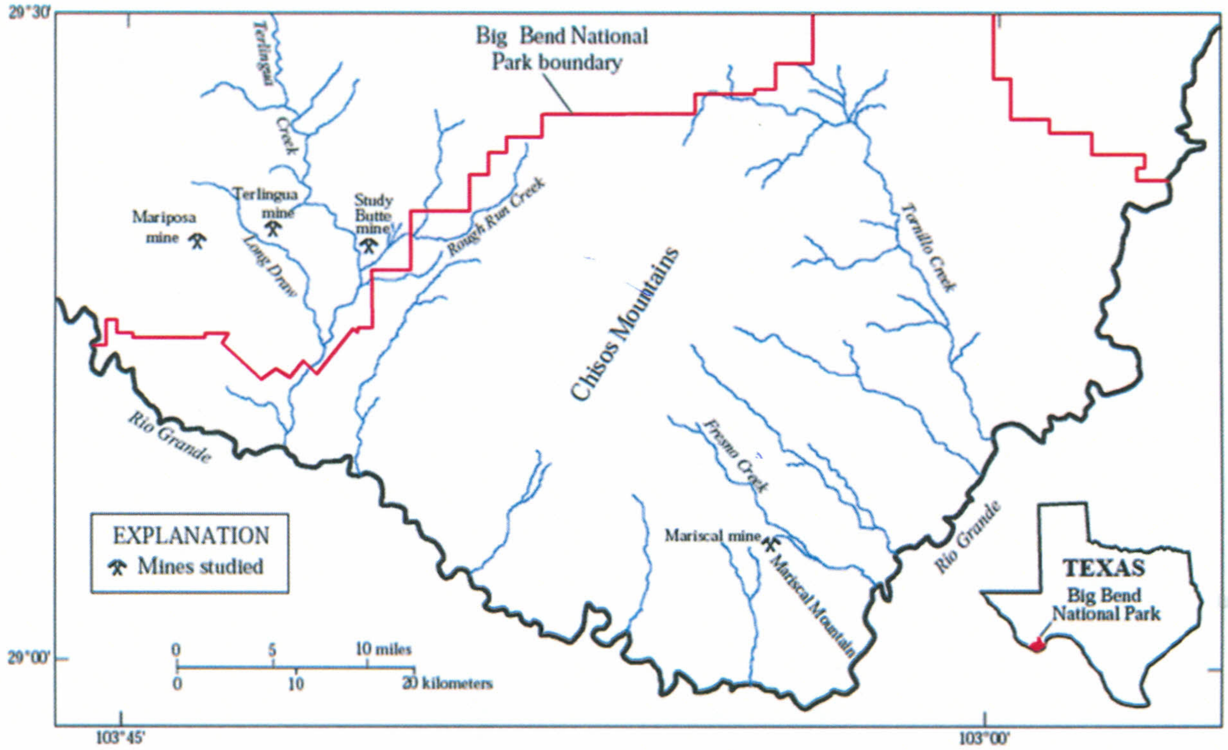

Figure 2: The Big Bend area of Texas and study area. U.S. EPA Circular 1327

Previous studies were conducted on the water, soil gas, and air on and around these mines. The water samples were taken at five separate locations around these mines: (1) near mercury mines, (2) the Rio Grande, (3) uncontaminated baseline streams, (4) hot and cold springs, and (5) wells in the Big Bend region (EPA Circular 1327, 2008).

The water samples collected were far below the drinking water standard set forth by the Environmental Protection Agency (EPA) for mercury at 2000 ng/L (EPA, 2003) (Fig. 3). However, long-term low-level exposure to aquatic life in the Rio Grande is evident per the conducted studies.

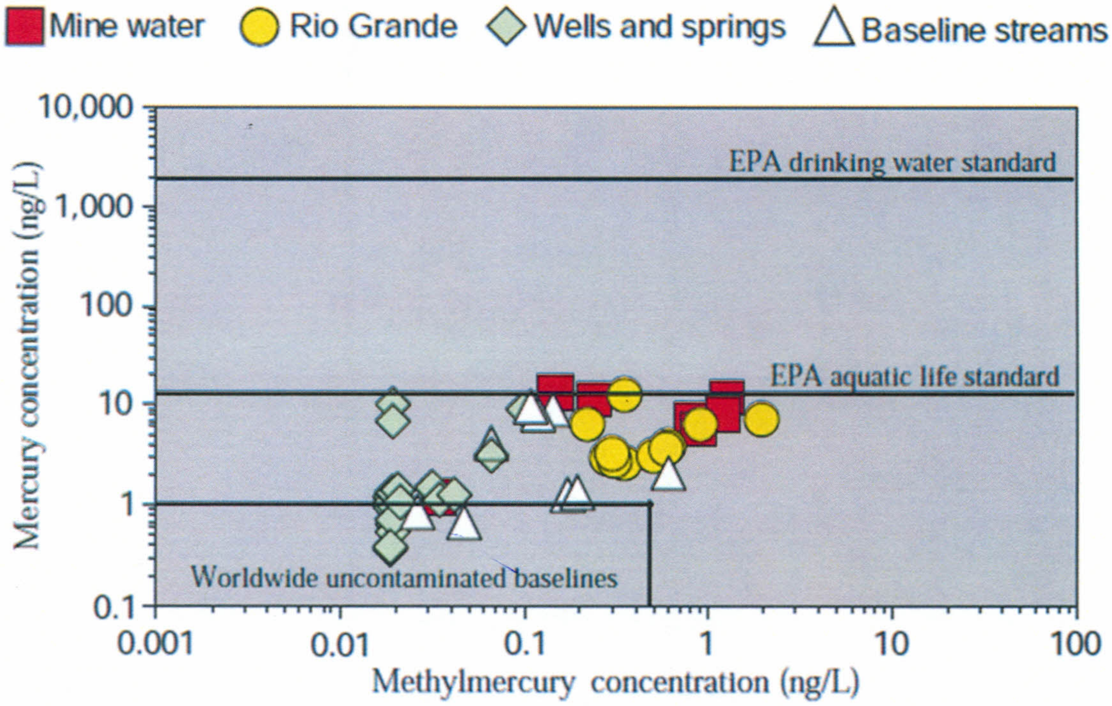

Figure 3: Concentration of mercury versus methylmercury in water in the Big Bend area collected around the Big Bend area. . U.S. EPA Circular 1327

The findings of the air and soil gas were taken from the Mariscal Mine and the old Chisos Mines at Terlingua. The range for emitted mercury from the mine waste is from 690 to 1,600 ng/m^3. However, readings from the Mariscal Mine brick retort are significantly higher at 18,000 to 21,000 ng/m^3 (EPA Circular 1326, 2008). These emission results of mercury flux are typical of similar mines worldwide (Ferrara et al., 1991) (Gustin et al., 1996) (Gray, 2003) (Fig.4).

The study conclusions are that concentrations of mercury and methylmercury are altogether low in stream sediment and water samples. The indication of the semi-arid climate in the region would restrict the transportation of methylmercury downstream to the most important ecosystem in the area, the Rio Grande (Gray et al., 2006).

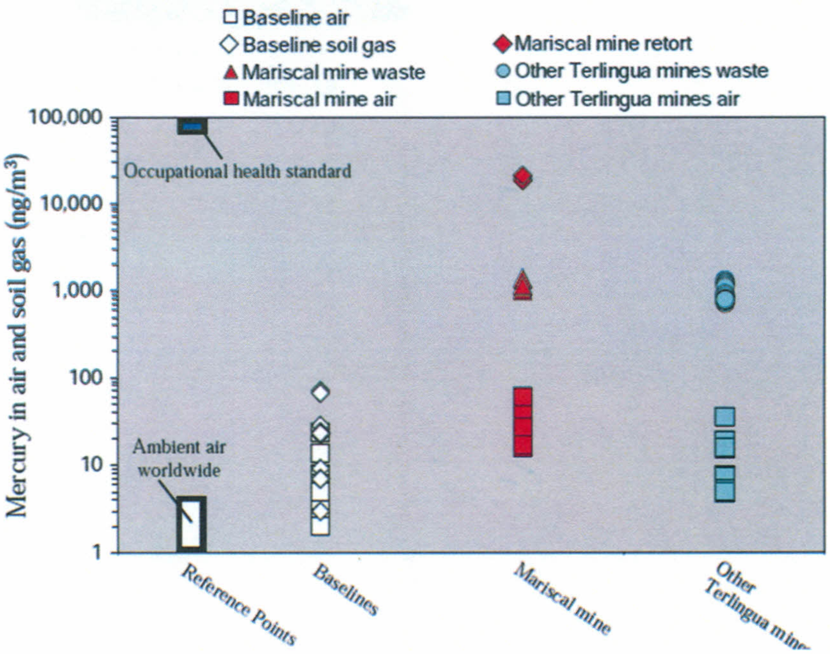

Figure 4: Mercury concentrations in air and soil gas collected from mercury mines in the Big Bend area and from uncontaminated baseline sites in the park. . U.S. EPA Circular 1327

Traditional remediation methods for soil are solidification/stabilization, soil washing/acid extraction, thermal desorption/retorting, and vitrification (EPA, 2007). These methods can be very costly even with facilities in close proximity to the contaminated site. Terlingua's remote location and expansive area would only exacerbate the costs of traditional remediation methods. In 2004, the EPA considered 63 hard-rock mining sites as priority superfund sites; Terlingua was not one of them. The costs associated with the cleanup for 2004 was estimated at 7.8 billion U.S. dollars (Government Accountability Office, 2004). As of 2013, the same 7.8 billion dollars is estimated at 9.61 billion in 2012 U.S. dollars, the cumulative inflation rate of 23.2% (U.S. Inflation Calculator). The Reclamation Act of 1977 provided funding for the Texas Railroad Commissions Abandoned Mine and Land Reclamation Program (AML) to clean up surface mines and close underground mine openings. As of 2001 the AML closed the 400 plus underground mine entrances and did nothing to address the contaminated land and spoils piles in the former Terlingua Mining District (Texas Railroad Commission, 2013).

Phytoremediation is the use of living plants and their microbes used in the removal or degradation of contaminants, either organic or inorganic, from the natural environment. Phytoremediation uses a plants natural process with their microbial rhizosphere flora to degrade and sequester organic and inorganic pollutants (Pilon-Smits, 2005). These plants can be native, non-native, in situ, ex situ, transposed, terrestrial, or aquatic. The pollutants can be highly toxic, carcinogenic, and while predominantly man made, being xenobiotic to all organisms. Phytoremediation is a cost-effective and minimally environmentally detrimental solution (Mendez and Maier, 2008). Phytoextraction is a sub category of Phytoremediation in which the metallophyte plant is used to remove and amalgamate the heavy metal contaminants from the soil into the plants aboveground leaves and branches (Fig. 5). After harvesting the plants, now rich in metals, these plants can be processed easily and safely by composting, drying, or ashing. A few extracted metals, such as cadmium, nickel, lead, and copper, can be reclaimed from the ash using safer and less environmentally damaging

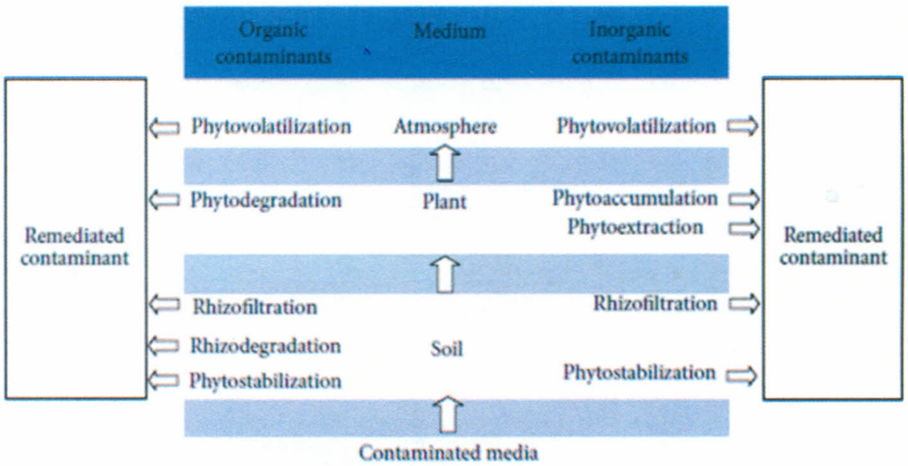

Figure 5: Phytoextraction from soil to the stems and leaves illustration. Tangahu et al., 2011

reclamation methods, generating recycling revenues (Garbisu and Alkorta, 2001). Phytoremediation may be the substitute to the less environmentally friendly traditional remediation methods. Traditional methods can destroy the biological constitution of the soil while changing the chemical composition and making it a worthless solid waste product (Hinchman et al, 1995).

There can be certain limitations to phytoremediation. Depending of the species of plant/bush/tree, phytoremediation can be a time consuming endeavor. Instead of a few weeks or months with a traditional remediation method, phytoremediation may take multiple growing seasons or years to remediate a site. Sites that pose great risk of heavy metal exposure to human populations and fragile ecosystems, phytoremediation may not be the remediation technique to be used (Tangahu, 2011). The best appropriate areas for Phytoremediation would be remote locations where there is a limited human interaction or soil contamination does not require immediate action (Salido et al, 2003).

Root contact with the contaminated soil is essential for the success of the phytoremediation technique. The plants must be able to extent their roots to the contamination zone within the soil. Many plants have a shallow root zone therefore deeper contamination zones would need a traditional remediation method (EPA, 2000)(Mwegoha, 2008).. In semi-arid climates, there are some plants that can tap into shallow aquifers with their roots.

There is a diverse array of native Chihuahuan desert plants growing near the tailings piles in the former Mining District. A few of the species include: sotol (*Dasylirion liophyllum*), four types of yuccas, Faxon yucca (*Yucca faxoniana*), beaked yucca (*Yucca rostrata*), soaptree yucca (*Yucca elata*), and torrey yucca (*Yucca treculean*), prickly pear (*Opuntia),* ocotillo (*Fouquieria splendens*), *Texas ranger (Leucophyllum frutescens*), Indian mustard plant *(Brassica juncea),* burro sage *(Ambrosia dumosa),* various types of grasses and flowers, and many other native plants (National Park Service, 2013). Of all the native plants in the Chihuahuan desert and most prevalent in the former Terlingua Mining District, that are the most promising in the phytoextraction of Hg from the soil, is the creosote bush (*Larrea tridentata*) (Fig. 6). The creosote bush naturally grows on the top, slope, and base of the tailings piles. Therefore, this plant is ideal to use for this study.

Figure 6: Creosote bush (Larrea tridentata) near Terlingua. http://cybertao.blogspot.com/2010_05_01_archive.html

The creosote bush is a perennial, drought-tolerant, evergreen shrub growing upwards to 13 feet (4 m) tall (Munz and Keck, 1959). The plant consists of friable stems or branches where leaves are denser towards the ends (Fonteyn and Mahall, 1981)(Munz and Keck, 1959). Due to the arrangement of the stems and leaves, sparse shade is provided during the day (Mares et al., 1977). The creosote bush is native to all the deserts of the American southwest and covers approximately 40% of the Chihuahua Desert (MacMahon, 1988). The common growth distribution is on bajadas, slopes, valley's, dunes, and in arroyos (Burgess and Northington, 1974)(Darrow, 1944)(Went and Westergaard, 1949), elevations up to 5,000 feet (1,524 m) (Kearney et al., 1960)(Munz and Keck, 1959). Growth occurs on calcareous, sandy, and alluvial soils that are dominated by a caliche hardpan at a shallow depth of about 3 feet (1 m) (Brown, 1982)(Gardner, 1951) (Gehlbach, 1967)(Haase, 1972)(MacMahon, 1988).

The root system of a creosote bush is normally comprised of a single taproot and multiple lateral or secondary roots. The taproot normally extends to a depth of about three feet (1 m) where it is then inhibited from further growth by caliche. The lateral or secondary roots reach an average length of 10 feet (3 m) and reach a depth of 8 to 14 inches (20-35 cm) which can also be inhibited by caliche (Fonteyn, 1981)(Singh, 1964). Other studies of the creosote bush state that the taproot, if not inhibited by caliche, can grow to a depth of 10 feet (3 m) (Mahall and Calloway, 1992).

The rainy season in the Big Bend area is May through October (National Park Service, 2013). While flowering normally occurs during the spring, summer, and fall rains, flowers can bloom during the winter months if enough rain is received (Ackerman et al., 1980)(Ackerman and Bamberg, 1974)(Barbour et al., 1977). Seed germination development appears in June through September (Ackerman, 1979) with a natural shedding of the leaves occurring in the fall (Barbour, 1968).

In previous studies, the creosote bush was found growing naturally on heavy-metal contaminated soils near a copper smelting plant near El Paso, Texas. The heavy-metal contaminates tested in these soils were copper, lead, cadmium, nickel, arsenic, chromium, barium, selenium, and zinc. The creosote bush was then analyzed and other metals were shown to be in the creosote tissue. These metals were mercury, strontium, selenium, and arsenic (Gardea-Torresdey et al, 1997). With Lead being the most difficult to absorb, translocate from the plants rhizoshere, into the stems, and leaves after absorption (Kabata-Pendias, 1989)(Koeppe, 1981)(Lepp, 1981). Heavy metals, in the terms of this prospectus, is any metal having a greater atomic mass than that of sodium as well as some other elements found around mine tailings such as arsenic and selenium (Gardea-Torresdey et al, 1997).

The creosote bush, being a perennial, is unique. During harvesting, many other plants such as Indian mustard (*Brassica juncea*) will need to be removed from the contaminated site and disposed, therefore needing to replant. Not the creosote bush. After the heavy metal or metals have been sequestered in the leaves and stems, the bush does not need to be removed from the contamination site. At set time intervals the leaves and stems are removed allowing the plant to survive. Harvesting can take place annually or bi-annually allowing for the continuation of phytoextraction of the contaminated soils. The collected leaves and stems are then properly disposed (Gardea-Torresdey et al, 1997). Gardea-Torresdey's research was not specifically testing for mercury however, mercury was found in the tissues of the creosote bush.

The purpose of my study is to use the creosote bush in the phytoextraction of organic mercury (methylmercury or MeHg) from a single tailings pile left over from the old Chisos Mine located in the former Terlingua Mining District in the Big Bend, Texas.

Methods

A single tailing or gangue pile will be selected for the study of the phytoextraction of Hg by the creosote bush. The selected study site is at geodetic location 29°19'06.87" N 103°37'23.49" W elev. 2890 ft. (Fig. 7) (Google Earth). Thirteen creosote bushes will be purchased from One Way Plant Nursery in Alpine Texas. Per

Figure 7: The former Chisos Mine tailings piles circled in black with study tailings pile circled in red. Google Earth, Terlingua Texas 2013.

Nancy, the owner, creosote bushes come standard in a five gallon container with the plant above the root ball an average of three feet tall and about one and a half foot wide. The cost is normally $20 to $25 per bush. Once the 13 bushes are purchased, each bush will then receive a unique number. A sample of leaves, stems, and fruit will then be removed from the specifically numbered bush and stored in an identical numbered container. The 13 samples will then be sent to an EPA certified lab for testing of heavy metals. The samples are to be tested for the normal heavy metals found in and around inactive and mines. These heavy metals are chromium, cobalt, nickel, copper, zinc, arsenic, selenium, silver, cadmium, lead, and of course mercury. Detailed testing of each bush prior to planting will give us a baseline for any metals already resident in the specific bush prior to planting. Note, most plants from nurseries will have delayed release fertilizers mixed in with the root ball soil. The water used for each plant during planting will also be tested for the same aforementioned heavy metals. Sul Ross State

University is located in Alpine therefore possible costs of testing of both bush and water could be minimized. If Sul Ross State University cannot provide testing services, then possibly The University of Texas at El Paso or a third party testing facility. A one gallon water sample will be taken from bush before each individual planting. If metals are not tested prior to planting in both the bush and water, skewed results could occur. Water will come from the shallow aquifer that is adjacent to Terlingua Creek and its tributaries which is also used by the residents of Terlingua. This water is somewhat saline and hard (Fallin, 1990).

The best time to plant the creosote bush is either in late winter or early spring. The hole to be dug should be twice as wide as the root ball of the bush. The depth of the hole should be no more than six inches (15 cm). The top of the root ball should be Level with the ground in to which you are planting. If on a slope, the top of the root ball should be level with lowest portion of the open dug hole. Prior to setting the bush in the hole, use your fingers to loosen the soil around the root ball in order to free the roots from their prior containment. Let the soil from the root ball settle in the hole. The loose soil around the root ball will allow the roots to expand and establish. Once the bush has been planted water the soil enough to saturate the ground (Nokes, 2001). March and April receive slightly over an inch of rain during each of these month, (National Park Service, 2013), therefore; the planting time will be after the first day of spring, March 20 and before April 1. These two months are the ramp up to the local areas rainy season. No more watering will be needed by the researchers.

The tailings pile selected for the research is approximately 100 ft. (30 m) in height and 320 ft. (97 m) around at its base. The tailings pile faces south with two gullies running down either sides, one facing west and the other facing east. There are multiple rills running down the face of the tailings pile from the northeast, around to the south, and continuing around to the northwest. The higher elevation of the local topography is connected to the tailings pile to the north.

The planting of the creosote bushes will be strategically placed on the tailings pile. Creosote bush numbering will be from top to bottom, left to right, and in five rows. Row 1 is the very top and will only have one bush in the row. Row 2 is below row 1 and will contain bushes 2 and 3. Row 3 is below row 2 and will contain 4, 5, and 6. Row 4 is below row 3 and will contain bushes 7, 8, 9, and 10. Row 5 is below row 4 and will contain bushes 11, 12, and 13 (Fig. 8). There are naturally growing creosote bushes on the tailings pile so some of these may need to be removed in order for us to conduct our

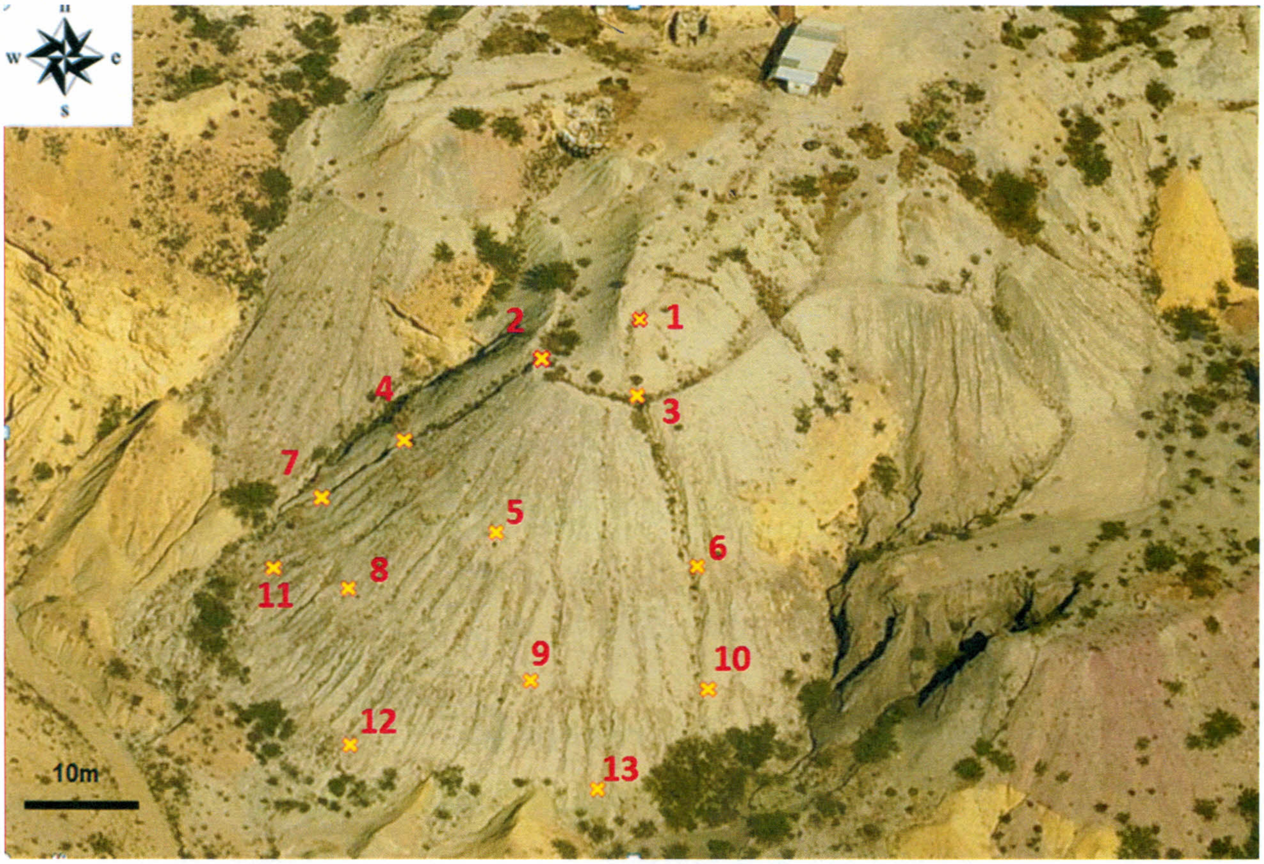

Figure 8: Tailings pile study site with creosote planting location designations and numberings. Bing Maps, Birdseye view 2013.

research in these locations. Locations 2 and 3 will most likely be the locations for the removal of current creosote bushes. The location number will correspond to the creosote bush number.

Extreme caution will be used in the planting on the slope of the tailings pile. The disturbance of the tailings from above one or more of the test bushes could release excess soil gas and transportation of mercury during rain events. To keep the disturbance to a minimum on bushes 2, 4, and 7 we will enter the tailings from the west. For bushes 3, 6, and 10 we will enter the tailings form the east. Bushes 8, 9, 11, 12, and 13 we will enter the tailings from the bottom of the pile. For bush 5 we will enter the tailings pile at an angle from between bushes 9 and 12. Bush 1 can be accessed from the top of the tailings pile.

The measurement of the height and width of each bush will be taken prior to planting. The growth rate of each bush will also be tracked throughout the research time interval. The collection times for stems, leaves, shallow rootlets, possible fruit as well as the measurements of bush growth will be taken every three months starting after the initial planting at the end of March. Collection times will be at the ends of June, September, December, and March of the following year. Collection during the months of June and September is during the rainy season while December and March are considered the dry season (National Park Service, 2013). The research time interval will be two years with collection and measurements to be conducted every three months. Testing and measurement intervals of every three months may seem excessive but we need to measure Hg uptake and growth rate, if any, during both the dry and wet seasons. Blooming occurs during the rainy season but will also bloom during the dry season when adequate rains arrive. Therefore, the possible growth of the bush and uptake rates could be continual throughout the year. Testing and measurement of the bush during the dry season will measure the uptake differential between rainy and dry seasons as well as growth dimensions of the bush. A two year timeframe will allow the creosote bush to establish its taproot as well as its secondary lateral roots. The grow depth of taproots and secondary lateral roots of bushes 1 through 10 should not be hindered from a caliche' layer. However, the possibility for bushes 11 through 13 having a caliche' layer restriction is higher due to the placement of the bushes at the bottom of the tailings pile.

Results from the initial testing of the bushes prior to planting, during each collection interval, and the end results will be analyzed. The comparisons between initial testing results and ending results of each plants uptake rates will be compared. This will reveal the total amount of Hg uptake as well as any other metals tested for that may be present in the tailings. Analyzing the results from each bush at the three month interval will give a progression uptake rate through the seasons. Wet seasons can be compared to each other and to the dry season to verify if the uptake rate is higher in the rainy season. The growth height and width can be compared to uptake rates in both wet and dry seasons. A comparison of precipitation rates, growth rates, and uptake rates can help in determining if the creosote bush is a viable option to traditional remediation methods.

Conclusion

Scientists agree Hg is toxic to all biota. The remediation of man introduced Hg pollutants in our soils, water, and air is imperative. Traditional remediation methods are costly and even exacerbated in remote locations. This research proposes a non-traditional remediation method through a relatively new and promising method, phytoremedition. The creosote bush thrives in semi-arid and arid regions of the world which makes it a suitable species for the phyoremediation of the former Terlinqua Mining District. This bush has shown to extract various heavy metals from desert soils into its tissues and sequester these metals. Unlike other desert plants that extract heavy metals from the soil, such as the Indian mustard, the creosote bush during harvesting, does not need removed from the soil. The leaves, stems, and sometimes fruit can be removed leaving the core of the bush in the ground to continue removing the heavy metals from the soil.

The phytoremediation of Hg using the creosote bush is not without its problems in this region. The creosote bush provides a home for multiple specialist insect species. The flower provides the nectar for 22 species of bees and is the sole food supply for the creosote bush grasshopper (Bootettix argentatus) (Pavlik, 2008). Will these species be affected by the phytoremediation of Hg by the creosote bush, only further research will answer the question?

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