

# History, Geochemistry and Environmental Impacts of Contaminants Released by Uranium Mining in South Texas

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## Abstract

In 1954, uranium was discovered in the Nueces River watershed near Tordilla Hill, western Karnes County. Uranium discoveries expanded (northwest and south) to an estimated  $U_3O_8$  resource of 100,000 tons. From 1960 until 1983, uranium mining within the Nueces (and adjacent) watersheds generated large-scale landscape impacts including open-pit excavation, overburden pile accumulation, drainage course alteration and ground and surface water contamination. By 1980, the Texas Gulf Coast ranked 3<sup>rd</sup> in production of  $U_3O_8$ , (3 million lbs/yr) in the United States.

Uranium mining liberated arsenic (As), molybdenum (Mo), selenium (Se) and vanadium (V) to the nearby environment via wind and water. Pit dewatering impacted nearby drainages; stockpiled tailings eroded and acidic processing wastes leached from unlined mine pits. Livestock, grazing near uranium mines, were poisoned by excess molybdenum. Although contaminants released from uranium mines have adversely impacted nearby areas, the effects of mining have not been evaluated at the watershed scale.

It is likely that U and U-associated mining impacts have been transported away from the immediate vicinity of mines. Uranium, As, Mo, Se and V exhibit high geochemical mobility in oxidizing water and are immobilized in reducing waters. In New Mexico, these contaminants have been shown to migrate from uranium mines via (oxidizing) surface water and to accumulate in (reducing) organic-rich fluvial sediments up to 100 km downstream. A GIS is being used to assemble information regarding the location of pits, tailings piles, discharge points, surface water drainages and wetlands in the Nueces watershed. These data layers are directing sample collection aimed at revealing the magnitude of uranium mining impact.

The talk will present the history of uranium mining in the Texas Gulf Coast, a review of the geochemistry and environmental significance of As, Mo, Se and V and preliminary data from sampling and analyses.

## INTRODUCTION

Texas was the third largest producer of uranium oxide ( $U_3O_8$ ) in the United States in the late 1970s and early 1980s. Mined exclusively from the Gulf Coastal Plain (Figure 1), Texas produced 3 million lbs. of  $U_3O_8$  in 1981 (Texas Railroad Commission, 1983).

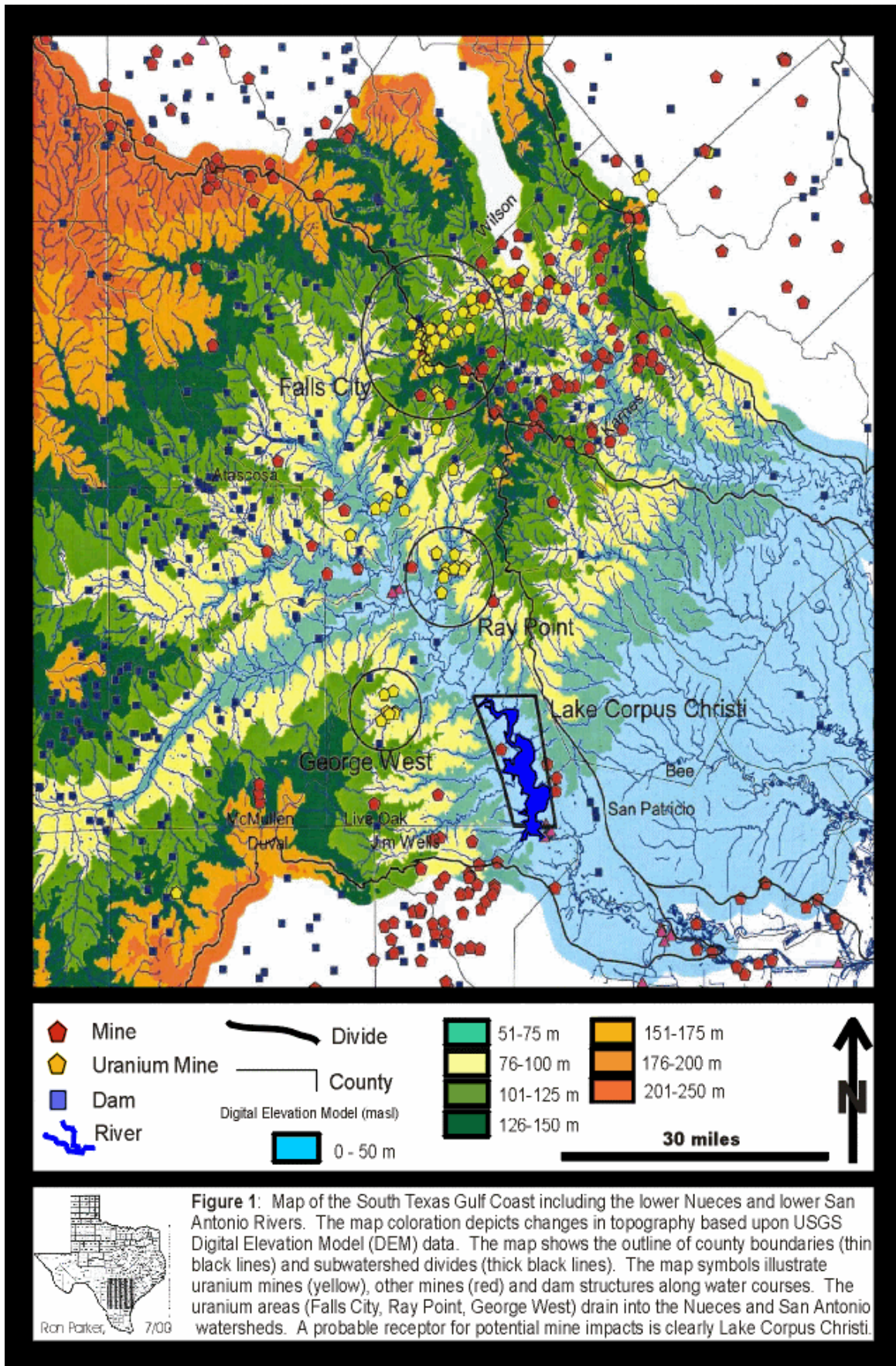
One of the most surprising and interesting characteristics of the uranium mining history of South Texas is the degree to which this history is presently invisible. Very few people residing along the Texas Gulf Coast are aware of the societal, economic and environmental impact that uranium mining has had upon South Texas. The uranium mining industry simply has not been a dominant presence in the consciousness of Texans, a phenomenon likely because uranium mining in South Texas was a very large-scale activity hosted in a very sparsely populated region. As a result, only a small number of individuals were directly affected by mining operations. Most of the mining region remains sparsely populated, where the vestiges of mining have been further removed by a combination of regulated restoration efforts and the rapid recolonization of the perturbed landscape by native vegetation.

An illustration of the “invisibility factor” is found -or, more accurately, is NOT found - on the 1987 edition of the “Mineral Resources of Texas” map published by the Bureau of Economic Geology (Garner et al., 1987). The map lists all manner of mining localities and the distribution of mineral commodities. In spite of the large geographic range in which uranium has been found in Texas and the significant history of uranium mining here, mention

of uranium is conspicuously absent in any form on the map.

As we proceed into the 21<sup>st</sup> century, the population of the Texas Gulf coastal zone is projected to experience an enormous growth surge with the subsequent impact on water resources (Kufus, 2000). In order to protect water quality and ecosystem functioning along the Texas coastal margin, scientists, educators, legislators and policy makers must have a better understanding of the fate and transport of toxic trace elements across the coastal plain. To this end, a large effort has been initiated to catalog and characterize all of the point and non-point pollution sources within the Nueces River watershed. In the report on sources of nonpoint pollution in the lower Nueces watershed prepared under the auspices of the Corpus Christi Bay National Estuary Program (CCBNEP) (Baird et al., 1996) there is no mention of the potential nonpoint source contribution of trace elements liberated by the dispersion of uranium mining wastes. Uranium wastes seem to have been missed by CCBNEP programs as potential sources of non-point pollution in spite of the occurrence of a major uranium mining loci along Sulphur Creek in the Ray Point area of Live Oak County. Again, the uranium mining history seems to have vanished.

In spite of the invisibility of South Texas uranium mining, we contend that the legacy of mining and milling remains within the lower Nueces watershed. At present, the magnitude of uranium mining impacts is not known. Our research is aimed at characterizing the distribution of trace elements from South Texas uranium mines to more fully develop risk assessments. This paper presents the background of our investigative effort. The paper is



**Figure 1:** Map of the South Texas Gulf Coast including the lower Nueces and lower San Antonio Rivers. The map coloration depicts changes in topography based upon USGS Digital Elevation Model (DEM) data. The map shows the outline of county boundaries (thin black lines) and subwatershed divides (thick black lines). The map symbols illustrate uranium mines (yellow), other mines (red) and dam structures along water courses. The uranium areas (Falls City, Ray Point, George West) drain into the Nueces and San Antonio watersheds. A probable receptor for potential mine impacts is clearly Lake Corpus Christi.

organized into three sections. The first section reviews the history of uranium mining; the second reviews the geochemical behavior of the nonpoint-source contaminants released by uranium mining activities including U, As, Mo, Se, and V. The final section reviews documented and suspected environmental impacts of South Texas uranium. The information contained in this paper is the background foundational to an on-going investigation of the spatial and temporal distribution of trace elements released from the South Texas uranium-mining district.

### SOUTH TEXAS URANIUM MINING HISTORY

Because of uranium occurrence the southern Texas Coastal Plain has been defined by the United States Geological Survey (USGS) as the Gulf Coast Uranium Province (Finch, 1996). The Gulf Coast Uranium Province is large. It extends from the international border to Fayette County in the north and from the coastline to the outcrop of Eocene fluvial and marginal marine clastic rocks in the west (Galloway et al., 1979a; Finch, 1996). According to Finch (1996), a uranium province is "a large geologically and tectonically distinct region where substantial uranium is concentrated into clusters that each have a minimum of 500 tons of contained  $U_3O_8$ ." (p.3). The Gulf Coast Uranium Province is estimated to have a total resource magnitude of 100,000 tons of  $U_3O_8$  (much of which has already been removed by open-pit and in situ "leach" mining) (Finch, 1996). In the early 1980s, Texas was the 3rd ranking producer of uranium oxide ( $U_3O_8$ ) in the United States, accounting for some 3 million pounds of  $U_3O_8$  per year (Texas Department of Agriculture, 1991).

Uranium was discovered in 1954 near Tordilla Hill in western Karnes County by G. A. "Pat" Strodtman while conducting an aeroradioactivity survey as a means of finding petroleum. At nearly the same time, a private prospector (Mr. Ewers) found uranium-bearing minerals while looking for opalized wood near Tordilla Hill (Eargle et al., 1958). The discoveries set off a flurry of "intense exploration activity" by individuals and major oil and mining companies (deVergie, 1958). A mineral-prospecting guide published by the Bureau of Economic Geology entitled "Prospecting for Uranium in Texas" went into four printings in six months, distributing over 10,000 copies (Flawn, 1967). The "uranium boom" had captured the imagination of a large population of Americans seeking easy riches.

A series of aeroradioactivity flights were flown in the vicinity of Tordilla Hill to provide USGS geologists with a basis for more detailed ground-based study and to characterize the region before disturbance (Blanchett, 1979; Brown et al., 1961a; Brown et al., 1961b; Eargle et al., 1961a; Eargle and Moxham, 1961; Eargle et al., 1961b; Eargle et al., 1961c; Eargle et al., 1961d; Moxham, 1972; Trumbull et al., 1961). Over 500,000 feet of exploratory drilling was conducted across Karnes County, with an average hole depth of 80 feet (deVergie, 1958). Approximately 25 prospects were discovered along a 300 mile stretch of upper Eocene rocks from Fayette to Starr Counties within two years (Eargle et al., 1971; Flawn, 1967). By 1958, over 170 prospects were examined by AEC geologists (Flawn, 1967).

The initial ores were found to be within several feet of the surface and were easily extracted by backhoe and bulldozer. In 1957, several bulldozed pits were opened and approximately 8 tons of high-grade ore were removed (Eargle et al., 1958). At that time, the ore was shipped to processing mills in the Grants Uranium District in New Mexico. Largely due to the prohibitive cost of shipping raw ore to New Mexico, uranium mining proceeded at a relatively slow pace in South Texas for several years (deVergie, 1958).

As a result of drilling, it was discovered that Texas uranium

mineralization occurred as "roll front" deposits similar to those described previously in Wyoming. A roll front consisted of a crescentic zone of mineralization surrounded by an oxidized updip (concave) and a reduced downdip (convex) configuration (Figure 2) (Goldhaber et al., 1978; Reynolds and Goldhaber, 1978). Sea-level flux and lateral migration of depositional environments during the Eocene resulted in the interstratification of sand-rich and clay-rich materials along the coastal margin of South Texas. These strata were perfect for developing the conditions that led to formation of roll-front deposits due to the regional southeast dip created by subsidence (Galloway et al., 1979b). The source of uranium has been widely cited as being the Oligocene Catahoula Formation, which contains an abundant rhyolitic volcanoclastic component thought to derive from the Sierra Madre Occidental of Mexico (Ledger, 1981). Unaltered portions of the ash have been found to contain a minimum of 7 parts per million (ppm) of uranium (Galloway and Kaiser, 1980).

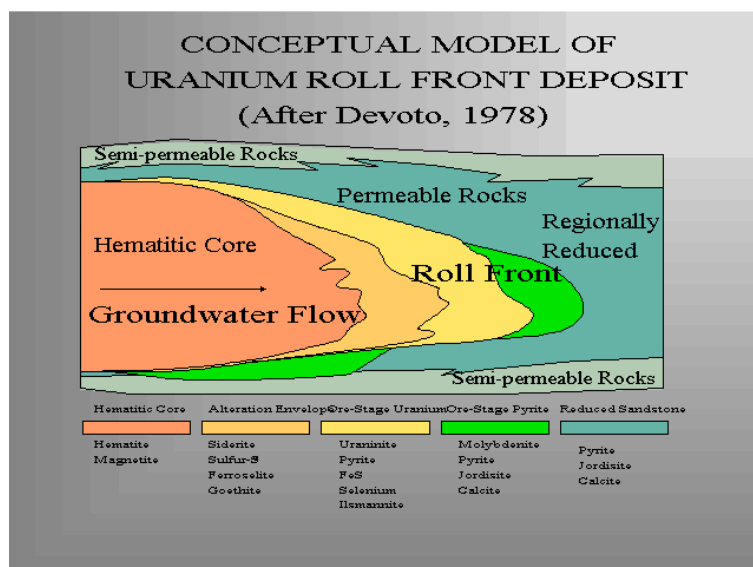


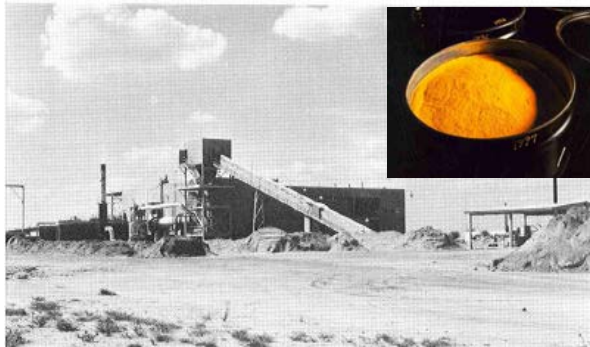
Figure 2: Schematic Roll Front; oxidizing to the left; reducing to the right. Source: Unknown Internet

Roll fronts develop as uranium and trace elements are solubilized by weathering of ash and carried downdip by oxidizing ground water in fossil aquifers (Galloway, 1977; Galloway and Kaiser, 1980). Dissolved oxygen is consumed along the flowpath by reaction with aquifer reductants including organic matter, pyrite and  $H_2S$  gas leaking from down-to-basin faults (Bomber, 1980; Miller, 1979). With decreasing redox, trace elements are progressively removed from ground water by precipitation of insoluble phases. Uranium, As, Mo, Se and V undergo reduction at differing values of Eh, which results in As, Mo, Se and V zonation in the vicinity of the uranium ore (Galloway, 1982; Henry and Kapadia, 1980). Zonation, likened to chromatographic separation, creates selenium enrichment updip, and molybdenum enrichment downdip of the uranium (Harshman, 1974).

The first commercial endeavor mined 100,000 tons of 0.2 % uranium ore removed from the Nuhn lease in 1959 and 1960 by Climax Molybdenum Company (Eargle et al., 1971). In the 1950s and early 1960s, all uranium was mined and processed by private firms for the United States government through the Atomic Energy Commission (AEC). The AEC used the uranium for national defense and research activities—dominantly nuclear weapons production (United States House of Representatives, 1996). It was widely known that the AEC was interested only in the richest ore; ore of lesser concentration than 2 pounds  $U_3O_8$  per ton was not

processed (Manka, 1998).

In April 1961, a uranium-processing mill was constructed by Susquehanna Western Inc. (SWI) in the Deweeseville area of westernmost Karnes County (**Figure 3**). This mill, originally designed to process 200 tons of ore per day, was enlarged in 1967 to a capacity of 1000 tons per day (Eargle et al., 1971). The mill used a sulfuric acid leach - countercurrent, decantation, and solvent extraction process for extracting the uranium. Uranium removal efficiencies were about 85% at the SWI plant (Manka, 1998). Before the mill closed in 1973, it had processed 2.5 million tons of ore averaging 0.16%  $U_3O_8$ . The AEC was sold more than 700 tons of  $U_3O_8$  concentrate (yellowcake) derived from the mill (USDOE, 1995a).



**Figure 3:** Susquehanna Western Mill – Deweeseville (1961-1970). Inset: 55-gallon drum filled with yellowcake (uranium oxide). Source: Eargle and others, 1971

The SWI processing mill completed a contract with the AEC in 1965 and promptly closed. This lull in the industry lasted only a few months as a great surge in the demand for uranium attended the development of new technology increasing the efficiency of nuclear power plants (Flawn, 1967). SWI contracted with the government of West Germany to supply uranium to nuclear power plants (Eargle et al., 1971). Thereafter, the nuclear power industry created a boom for uranium that lasted until the Three Mile Island nuclear accident in 1979.

In 1967, a second uranium district was discovered near Ray Point in Live Oak County. SWI constructed a second processing mill near Ray Point to handle the ores from these mines (**Figure 4**). The Ray Point mill opened in 1970 and treated raw ores with an alkaline leach process to extract uranium due to the high carbonate content of the host Oakville sandstone sediments (Eargle et al., 1971). Ray Point district roll fronts were different than elsewhere because the uranium was reduced by hydrogen sulfide gas that leaked along down to basin faults Galloway, 1980.



**Figure 4:** Susquehanna Western Mill – Ray Point. Source: Eargle and others, 1971

As a result of drilling exploration, deeper unoxidized ore deposits were located in southwest Karnes County in 1963 (Galloway et al., 1979a). In 1969 Tenneco became the second company to actively mine uranium in South Texas by opening up these deeper mines and sending ore to the enlarged SWI plant at Deweeseville (Eargle et al., 1971). In late 1973 Continental Oil Company and Pioneer Nuclear Corporation developed the Conquista Project. The Conquista partners constructed another processing mill with a projected yield of 1750 tons per day approximately 1 mile from the initial SWI mill (**Figure 5**) (Eargle et al., 1971).



**Figure 5:** Conquista Project Mill. Source: RRC

In 1975, Solution Engineering Inc. (SEI) purchased the property from SWI. SEI extracted the spoil piles and tailings that had initially been “high-graded” by the AEC. SEI developed a technique using shallow injection and recovery wells with ion exchange resins for stripping out the uranium remaining in the spoil and tailings. The injected solution was residual acidic process waters from one of the former mill ponds (USDOE, 1995a). This technology became the founding method that later evolved into full scale *in situ* leach mining that is practiced in several locations (i.e. Kingsville Dome) today.

Uranium mining was conducted by excavating extensive open pits into the ground to expose the ore. Some of these pits were more than a mile in length; many more were small surface excavations up to a few acres in size (Eargle et al., 1971). In the early days of uranium mining, the objective of acquiring only the highest ore grades reportedly resulted in the genesis of much larger open pits (Manka, 1998). Open pits required the removal of large volumes of material including original topsoil, barren overburden, overburden with slight enrichment of uranium and trace elements, termed protore, and ore material itself. The non-ore was stockpiled near the open pit whereas the ore was segregated onto an ore pad for temporary staging prior to transport to the processing mill. Early open pit mining was conducted by dump truck, resulting in loose, readily eroded spoil piles (Rotter, 2000). After several years of this excavation method, removing overburden was accelerated by using earthmovers (**Figure 6**).

The overburden (or, spoil) piles were built up in a sequential manner that resulted in a general “inversion” of the original stratigraphy. During stratigraphic inversion the first layers removed from the open pit were the first layers placed in the spoil pile. Thus, the previous “top” of the undisturbed stratigraphic sequence became the base of the spoil pile. For many of the open pit mines, the undisturbed stratigraphy consisted of a more heavily weathered and oxidized upper section and a less weathered, less oxidized (reduced) section with depth. This corresponded to a lighter coloration above and a darker coloration below. The stratigraphic inversion attending development of the open pit is thus most easily seen in spoil piles as the darker lower layers of the pit reside upon the lighter upper layers of the pit (**Figure 7**). Because the darker deeper material is more enriched in trace elements, placement of this material on top of the spoils pile accelerated trace element dispersion by erosive processes.



**Figure 6:** The use of earthmoving scrapers greatly accelerated the rate at which overburden was removed. Source: CONOCO.

Uranium tailings in South Texas and elsewhere are associated with potential environmental contaminants, mostly inorganics. These include, U, Ra, Th, Al, As, Co, Cr, Mo, Ni, NO<sub>3</sub>, Se, Pb, Hg, and V (Longmire, 1989; USDOE, 1992a). Uranium remains in tailings because the extraction efficiency of the milling process ranges from 80 to 95% (Manka, 1998). Radioactive elements that are the longer lived offspring of the <sup>238</sup>U and <sup>235</sup>U decay chains are also present in the tailings. These elements include <sup>238</sup>U, <sup>234</sup>U, <sup>230</sup>Th, <sup>226</sup>Ra, <sup>222</sup>Rn, <sup>210</sup>Pb, <sup>210</sup>Bi and <sup>210</sup>Po (Walker, 1989). Many of these constituents are leachable and are found as contaminants in ground and surface waters surrounding mines and mills, including those in South Texas (Bryson, 1988; USDOE, 1991a).



**Figure 7:** Inverted stratigraphy exposed in severely gullied spoil pile. This pile was created in 1972 and was restored in 1999. Erosion redistributed spoil to Mexican Hollow – Weedy Creek – Atascosa River – Nueces River for 27 years. Photo by R. Parker

Wastes produced by the processing mills were stockpiled in large tailings ponds where volume reduction via evaporation was promoted (Figure 8) Each of the four processing mills had a large scale tailings pond except the first mill at Deweeseville. Tailings wastes at the SWI-Deweeseville mill were stored in three smaller ponds or were pumped back into four abandoned open pits. One of the open pits was more than a mile from the processing mill (USDOE, 1995a). As the open pit mines were not lined, processing wastes infiltrated with adverse impacts to ground water quality.

In 1975, the Texas Legislature promulgated the Texas Uranium Surface Mining and Reclamation Act, which established the Surface Mining and Reclamation Division at the Railroad Commission of Texas (Texas Railroad Commission, 1982). The Act established a regulatory framework for mitigating the environmental destruction resulting from uranium mining activities via open pit mining. After passage of the Act, all responsible parties engaged in uranium mining operations were required to restore the open pit mine locations according to specifications designated in the law. Reclamation, in a general sense, required destruction of the pit high walls, placement of the most radioactive material back into the open pit, replacement of protore and spoil back into the mine pit, resloping and terracing the landscape to conduct surface water runoff into the open pit, applying topsoil cover and revegetating the site.



**Figure 8:** Conquista Project 245 acre Tailings Pond. Source: RRC

Operation of uranium mining enterprises after the law was enacted required permits that stipulated restoration activities with demonstration of the necessary financial resources to complete reclamation. Every open pit mine that was exploited after passage of the Act has been restored as of the present. In most instances, restoration left gently sloping spoil piles that surround a pit lake. Although some early restoration efforts were completed without leaving open water, it is now policy that reclamation be completed in such a way as to leave open water. Open water is highly prized in the semi-arid summers of South Texas and pit lakes are commonly used by landowners for watering livestock, for recreational boating and for fishing (Figure 9). Although no references have been found in the open literature, the Bureau of Radiation Control at the Texas Department of Health has certified most of the restored ponds as producing fish that are safe for human consumption (Meyer, 1999).

While open pit mines after passage of the Act have been restored, the same is not true of mines that were exploited prior to the passage. These mines, many of which were abandoned after removal of ore, are currently undergoing restoration closure by the Abandoned Mine Lands (AML) section of the Division of Surface Mining and Reclamation at the Texas Railroad Commission. The AML has catalogued 18 “pre-law” locations and has completed restoration on approximately 10 of them (Brandt et al., 1994). Restoration activities generally take about a year per mine and are projected to achieve completion by the year 2007 (Brandt, 2000).

In spite of the fact that abandoned mines are being restored, they still must be considered as sources of contaminants to the environment (Figures 7 and 10). During the operational lifetimes of the mining, spoil piles were exposed to erosional degradation. Many of the abandoned mines exhibit spoil piles that are deeply

dissected by erosional gullies (**Figure 7**). Some of these spoil piles have been sources of erosional debris (and associated contaminants) for up to 40 years.



**Figure 9:** Oblique aerial photo of the West Dzuik Pit in 1988 which was restored as a part of the Conquista Project. Note the vegetated spoil piles. This pit is a source of fish eaten by local residents. Source: RRC

In the United States, the environmental problems caused by releases of radioactivity and other chemical elements from abandoned uranium tailings piles was recognized as a hazard to human health and the environment. Consequently, in 1978, the United States Congress passed the Uranium Mill Tailings Radiation Control Act (UMTRCA) (42 USC § 7901 *et seq*) (Public Law 95-604), administered by the Department of Energy (Representatives, 1996a; USDOE, 1995a). As a part of UMTRCA, the Uranium Mill Tailings Remedial Action (UMTRA) Project was initiated. The directives of UMTRA identified 24 priority uranium tailings sites for remediation with an estimated budget of 900 million dollars (Deutsch, 1986; U.S. House of Representatives, 1996; USDOE, 1997c).



**Figure 10:** Aerial photo of the Weddington Open Pit in 1988. This mine was initiated in 1972 and was restored in 1999. Note the eroding spoil piles. The pit lake is approximately 3000 feet long. Source: RRC

The tailings from uranium ore processing at the SWI Deweeseville mill were selected for inclusion in UMTRA program

clean-up efforts. Known as the Falls City UMTRA site, this is the only UMTRA program site in Texas. It has the dubious distinction of having the largest volume of contaminated tailings (5,764,000 yd<sup>3</sup>) of any of the UMTRA sites (USDOE, 1997c). The Falls City UMTRA site consisted of the seven different uranium mill tailings impoundments (three ponds and four previous open pit mines). To date, the remedial efforts at the Falls City site include the consolidation of the tailings from the seven areas into one large stabilized, lined disposal cell. The disposal cell is physically located at the exact site of the SWI - Deweeseville processing mill. The dimensions of the tailings repository are 2200 feet by 2600 feet covering a footprint of 123 acres (**Figure 11**). This effort was completed in 1994 as a final cost of 38 million dollars (USDOE, 1998b).



**Figure 11:** UMTRA Tailings Repository from perimeter fence. Inset: View of pile from a distance of a mile to the south. Photos by R. Parker

This location was associated with ground water contamination to levels 1500 times EPA standards (Blount et al., 1992; Texas Senate, 1989). Unabated leaching of spent processing liquors introduced large volumes of concentrated sulfuric acid and entrained contaminants into the shallow ground water system. The rate of ground water contaminant transport was likely accelerated by cross-contamination through unplugged exploration drill holes. In the early days of uranium resource evaluation, thousands of feet of drilling and coring were conducted within a comparatively small area. In the 1950s and 1960s there were no environmental provisions for properly abandoning drillholes.

Ground water monitoring associated with the UMTRA program has revealed the presence of two extensive ground water contaminant plumes in the subsurface. These plumes are characterized by extremely low pH (related to the initial acidity of the sulfuric acid leach wastes) and abundant concentrations of contaminating inorganics (Al, As, Cd, Mn, Mo, Se, SO<sub>4</sub><sup>2-</sup> and U) (USDOE, 1995a). According to the USDOE, ground water contamination at the Falls City site has not resulted in surface water impacts as the result of ground water discharge to tributary drainages of the Atascosa (Nueces) and San Antonio Rivers (Texas Senate, 1989; USDOE, 1987). An absence of ground water discharge has not been adequately demonstrated in the opinion of the authors.

By 1989, more than a decade of delays by the USDOE initiating ground water cleanup at the Falls City site prompted the 71<sup>st</sup> Texas legislature to review the adequacy of extant state regulations governing the management of uranium mill tailings. As a consequence of the review, it was determined that there were serious deficiencies in the regulation of uranium mill tailings, which

resulted in a significant state liability. Additionally, it was determined that the serious “breach of acceptable regulatory practice” undermined the credibility of the Texas legislature to make and enforce regulations (Texas Senate, 1989). A significant finding of the Legislative report was that, because of permit abuses, industry auto-compliance and lack of legislative control, a significant contamination problem was likely at the uranium mill tailings sites in Texas.

Initial plans to clean up the ground water at the Falls City site resulted in preparation of a number of remediation options, the most favored having an estimated cost of 130 million dollars. Recently, the matter of ground water clean up has been rendered moot by the “Finding of No Significant Impact” (FONSI) of uranium tailings upon the ground water system at the UMTRA site. This finding was founded upon the determination that the native ground water initially present within the aquifers beneath the UMTRA site was already sufficiently degraded by natural processes as to render it unfit for human consumption or other use (USDOE, 1997c). Rendering the FONSI saved taxpayers approximately 130 million dollars in projected clean up costs.(USDOE, 1998b).

The active mining of uranium by open pit methods was brought to an end by the fall in the price of  $U_3O_8$ . In the 1960s,  $U_3O_8$  commanded about \$8 per pound. By the late 1970s, this had risen to a value of approximately \$44 per pound. After the accident at Three Mile Island in 1979, orders for new nuclear reactors were scrapped. This, in concert with the discovery of additional uranium resources in Australia and Saskatchewan, led to the precipitous decline in the price of  $U_3O_8$  and the failure of the industry in South Texas. Since cessation of the open pit mining operations, all activity in the open pit mining area has focused on restoration and closure.

#### GEOCHEMISTRY OF U, AS, MO, SE AND V

Uranium associated trace element enrichment in South Texas is due to the crudely similar geochemical behavior of U, As, Mo, Se and V (Harshman, 1974). Like uranium, As, Mo, Se and V are characterized by high geochemical mobility in oxidizing, neutral to alkaline waters, and low mobility under reducing conditions due to precipitation, complexation or sorption reactions (Henry and Kapadia, 1980).

Uranium forms the uranyl oxycation ( $UO_2^{2+}$ ) whereas As, Mo, Se and V are most stable as the oxyanions arsenite ( $AsO_3^{3-}$  or  $As(OH)_3$ ) or arsenate ( $AsO_4^{3-}$  or  $H_2AsO_4^-$ ), molybdate ( $MoO_4^{2-}$ ), selenite ( $SeO_3^{2-}$ ) or selenate ( $SeO_4^{2-}$ ), and vanadate ( $VO_3^-$ ) respectively (Anderson and Bruland, 1991; Hem, 1989; Pais and Jones, 1997). The oxyanions of As, Mo and Se are stable as uncomplexed ions (Tanizaki et al., 1992). Additionally, oxyions have a strong tendency to sorb to particulate matter, especially ferric and manganic oxides (Ames et al., 1983c), clay minerals (Ames et al., 1983a; Ames et al., 1983b; Batson, 1994) and humic and fulvic acids (Nakashima et al., 1984; Szalay, 1964).

It is well-known that trace elements occur in natural water systems in a variety of forms including individual hydrated ions, ion pairs, complexes with inorganic or organic ligands, colloidal complexes and as adsorbates to varying size fractions of particulate matter (Drever, 1988; Hem, 1989; Tanizaki et al., 1992). Elemental form and bioavailability are controlled by speciation. Speciation is in turn controlled by the master variables pH and Eh and other characteristics of the aqueous solution chemistry such as ionic strength (Stumm and Morgan, 1996).

**Uranium:** Uranium, (atomic number 92, atomic weight 238.0), is the heaviest naturally-occurring element and is the originating element (as both  $^{238}U$  and  $^{235}U$ ) for two of the three

radioactive decay chains that occur in nature (Walker et al., 1989). Uranium occurs in the 4+, 5+ and 6+ oxidation states; 4+ and 6+ being the most common. In oxidizing near surface environments, the highly mobile hexavalent ( $U^{6+}$ ) form is predominant (Langmuir, 1978a; Langmuir, 1978b; Langmuir, 1997). The uranyl cation ( $UO_2^{2+}$ ) predominates at low pH; the double uranyl dicarbonate complex  $UO_2(CO_3)_2(2H_2O)^{-2}$  is dominant in bicarbonate waters at a pH above 5 and the uranyl tricarbonate complex  $UO_2(CO_3)_3^{4-}$  dominates above pH (Langmuir, 1978b). Uranium dicarbonate and tricarbonate complexes are readily fixed by humic substances (Turner-Peterson, 1985). Advective transport of solubilized uranium is facilitated by strong sorption to clay minerals and micas (Ames et al., 1983a; Ames et al., 1983b; Batson, 1994), complexation with humic and fulvic acids (Nakashima et al., 1984; Szalay, 1964) and complexation and co-precipitation with other metals and colloidal particles including amorphous ferric oxyhydroxides (Ames et al., 1983c) and manganese oxides (Boyle, 1978). According to Boyle (1982), organic wetland deposits “are commonly enriched in thorium and uranium...” (p. 328). “Uranium shows little preference with respect to enrichment in particular varieties of organic bodies. All types of bogs, muskegs, marshes and wet-site mucky areas may act as collectors of uranium” (p. 329).

In South Texas, uranium has been measured in soils and stream sediments at concentrations ranging from background to 32,500 ug/kg (Henry and Kapadia, 1980). In surface water, uranium values from these areas have been found to range from background to 99 ppb (USDOE, 1995a). Uranium concentration in AML mine pit lakes ranges from below detection at 1 ug/L to 4,500 ug/L (Brandt and others, 1994).

**Arsenic:** Arsenic, (atomic number 33, atomic weight 74.92), is a toxic metalloid belonging to Group VA on the Periodic Table (Bello, 1988). The geochemical behavior of As has been found to mimic that of its Group VA neighbor P, although As forms bonds with a higher ionic character (because it is more metal-like) (Baird, 1995). Arsenic compounds coexist with those of phosphorus; As is a natural contaminant in phosphate deposits (Baird, 1995). Pentavalent As may substitute for phosphorus in metabolic processes resulting in disruption of oxidative phosphorylation and a blockade of adenosine triphosphatase (Anke et al., 1997). The well-known characteristics of As oxides as poisons stem from this interaction. Arsenic is a class A carcinogen on the basis of epidemiological studies in humans. It causes lung cancer when inhaled and skin cancer when ingested in drinking water (Krieger, 1999).

Zero-valent arsenic has a valence shell electron configuration of  $s^2p^3$ . Arsenic occurs naturally in the -3, 0, +3 and +5 oxidation states. The most common valence of As is +3 by loss of the three p electrons (Baird, 1995). The +3 oxidation state is the most toxic (Anke et al., 1997). Loss of the 5 valence shell electrons yields the +5 state (Baird, 1995).

Arsenic concentrations in aqueous solutions are controlled by equilibrium stability with solid mineral phases. In the uranium ore of South Texas, As is most likely to occur as a solid solution impurity with iron sulfides, as arsenopyrite ( $FeAsS$ ) or as the minerals Orpiment ( $As_2S_3$ ) or Realgar ( $AsS$ ) (Henry and Kapadia, 1980). In sulfur poor systems (i.e. most fresh waters) the dominant solid-phase species are adsorbed species.

Both inorganic and organic forms represent arsenic in natural systems. Reductive biomethylation of As produces a number of organoarsenicals including monomethylarsenic acid, dimethylarsenic acid, dimethylarsine and trimethylarsine (Anderson and Bruland, 1991; Anke et al., 1997). In the environment, the different As species have stabilities that are controlled by pH and redox conditions. In oxidizing, alkaline to neutral waters, As occurrence is dominated by the oxyanions arsenite ( $AsO_3^{3-}$ ) or

As(OH)<sub>3</sub> or arsenate (AsO<sub>4</sub><sup>3-</sup> or H<sub>2</sub>AsO<sub>4</sub><sup>h</sup>). Arsenate, (As<sup>5+</sup>) is the most thermodynamically stable form under well-oxidizing conditions; while arsenite, (As<sup>3+</sup>) dominates in reducing geochemical environments (although slow transformation kinetics permits the presence of both species in either setting) (Cullen and Reimer, 1989).

The mobility and bioavailability of As is controlled, in part, by adsorption to solid surfaces. The most important natural sorbents for As oxyanions are ferric oxides, hydroxides and oxyhydroxides (Cullen and Reimer, 1989). The pH dependent charge on these Fe species favor sorption of the negatively charged As oxyanions at pH values corresponding to positive net surface charge. Favorable pH values vary depending upon the identity of the sorbent, ranging from 4.2 to 6.9 (hematite) and 5.9 to 6.7 (goethite) to 8.5 to 8.8 (amorphous Fe-hydroxide) (Langmuir, 1997).

Studies of the environmental fate of arsenic have generally reinforced the observation that As is preferentially sequestered in sediments under most natural surface conditions (Cullen and Reimer, 1989). In the Aberjona watershed near Boston, As (up to 2000 mg/kg) was found to concentrate in the bottom sediments deposited in shallow lakes, whereas total concentrations of As in the water column were approximately 3 ug/L (Hemond, 1995).

In uranium areas of South Texas, As has been measured in soils and stream sediments at concentrations ranging from background to 482,000 ug/kg (Henry and Kapadia, 1980). In surface water, As values from these areas has been found to range from background to 44 ppb (USDOE, 1995a). Arsenic concentration in AML mine pit lakes ranges from 5 ug/L to 33 ug/L (Brandt and others, 1994).

**Molybdenum:** Molybdenum, (atomic number 42, atomic weight 95.94), is an element that is both an essential micronutrient for plants and animals and a toxic contaminant (Reddy et al., 1997). Molybdenum forms bonds with a highly covalent character and, hence, is considered to be a soft acid. Molybdenum exhibits a range of stable oxidation states from +2 to +6, the most common being +6 (Johnson, 1997). In the "higher" oxidation states (+3 to +6), molybdenum has a very strong affinity for oxygen and oxygen containing groups (Smith et al., 1997). As a consequence of this tendency, molybdenum is almost always found in natural aqueous solutions as an oxyanion, dominantly molybdate (MoO<sub>4</sub><sup>2-</sup>) (Hem, 1989). Molybdate forms solid precipitates with a wide variety of cations; some of the most important minerals resulting include molybdenite (MoS<sub>2</sub>), ilsemanite (Mo<sub>3</sub>O<sub>8</sub>), powellite (CaMoO<sub>4</sub>), wulfenite (PbMoO<sub>4</sub>) and ferrimolybdate (Fe<sub>2</sub>O<sub>3</sub> • 3.52 MoO<sub>3</sub> • 10.4 H<sub>2</sub>O) (USEPA, 1979).

The biochemical interactions of molybdenum are complex and manifold. Among the most notable are the interactions of molybdoenzymes, which catalyze redox reactions involving a broad spectrum of substrates in plants and microorganisms (Johnson, 1997).

Although molybdenum may be found as a variety of species, in aqueous solution it is generally present as the dissolved molybdate (MoO<sub>4</sub><sup>2-</sup>) anion. Except for surface water that is very acidic or that has a high content of particulate ferric iron, Mo occurs in the form of a "truly dissolved, filterable species" (USEPA, 1979, p.25). Anionic molybdate is the form that is readily available for uptake by plants. Molybdate does not form aqueous complexes with major cations, but it does complex with humic and fulvic acids. Redox conditions exhibit a strong controlling influence on the mobility of Mo: it is soluble in oxidizing conditions as molybdate and insoluble in anoxic conditions as molybdenite. As a consequence, Mo is commonly found in association with accumulations of organic matter in reduced geochemical environments (Smith et al., 1997). Mo is subject to pH dependant

sorption to ferric and aluminum oxides and to clay minerals with a sorption maximum near the pK<sub>a</sub> of HMoO<sub>4</sub><sup>-</sup> of 4.23 (Goldberg et al., 1996). The sorption reactions of Mo with these solid phases is characterized as specific adsorption whereby the Mo forms covalently bonded inner-sphere complexes with the solid surfaces (Erdman, 1990; Goldberg et al., 1996; Reddy et al., 1997). In alkaline conditions, molybdenum is not strongly sorbed resulting in enhanced mobility.

There is evidence for Mo-induced disease among humans, especially in less developed countries. Near a dam in southern India, local residents developed *genu valgum* (knock knee), a crippling bone disease attributed to flooding-induced increases in soil alkalinity and resulting increases in Mo in sorghum (Agarwal, 1975). The EPA has recommended a drinking water guideline of 50 ug/L (USEPA, 1979).

There are few studies of Mo cycling in lakes. In one study, a pronounced fluctuation of Mo concentration was observed in the total water column of a small glacial kettle lake in Connecticut. Mo concentrations were observed to peak immediately after the meltout of winter ice. At this time, the lake water was isothermal with depth close to a minimum temperature of 4 °C. These conditions would have maximized the concentration of dissolved oxygen diffused into lake waters, a condition which may have resulted in liberation of Mo from lake bottom sediments (Cowgill, 1977).

In uranium areas of South Texas, Mo has been measured in soils and stream sediments at concentrations ranging from background to 1.12% (11,200,000 ug/kg) (Henry and Kapadia, 1980). In surface water, Mo values from these areas have been found to range from background to 80 ppb (USDOE, 1995). Molybdenum concentration in AML mine pit lakes ranges from 10 ug/L to 650 ug/L (Brandt and others, 1994).

**Selenium:** Selenium, (atomic number 34, atomic weight 78.96), occurs in the -2, +4 and +6 oxidation states and in a zero-valent elemental form (Sunde, 1997). Inorganic selenium in natural waters occurs as the two oxyanions: selenite (SeO<sub>3</sub><sup>2-</sup>) and selenate (SeO<sub>4</sub><sup>2-</sup>) (Anderson and Bruland, 1991; Hem, 1989). The dominant organic selenium compounds include dimethyl diselenide, which is a volatile gas (Conde and Alaejos, 1997).

Selenium toxicity in animals, especially livestock poisoned by seleniferous forage, has been known for many years (Brooks, 1998). In livestock, acute toxicity causes the "blind staggers" whereas chronic exposure results in "alkali disease" (Sunde, 1997). Discovery of excess mortality and congenital defects in wildlife at the Kesterson National Wildlife Refuge and Reservoir in 1982 in California's San Joaquin Valley spurred a large research effort into the geochemistry, biochemistry and environmental toxicology of selenium (Frankenberger and Benson, 1994; Frankenberger and Engberg, 1998; Jacobs, 1989; Severson et al., 1991). A dominant factor controlling Se toxicity is evapoconcentration (Engberg et al., 1998; Ong et al., 1995). In partial response to the contamination at Kesterson Reservoir, the USEPA has set the aquatic life chronic Se criterion at 5 ug/L (5 ppb) (Seiler, 1996)

Selenate is thermodynamically favored in oxidizing waters; selenite in mildly reducing waters and elemental selenium and selenide under strongly reducing conditions. Water and sediment redox conditions, as well as pH and salinity are dominant controls over the selenium speciation (Jayaweera and Biggar, 1996; Tokunaga et al., 1996; Zawislanski and Zavarin, 1996). When oxidizing waters carrying selenate undergo oxygen depletion, the selenate is slowly reduced to selenite and elemental selenium, which sorb to sediment particles or precipitates out. Elevated pH and salinity favor Se adsorption to clays and metal oxides (Mayland et al., 1991). Selenate is the most soluble and bioavailable form and is characterized by weak adsorption to particulates. Selenite is

immobilized by adsorption onto particulate matter, especially Fe-oxyhydroxides. Selenide and elemental selenium occur as insoluble compounds in acidic, reducing and organic matter-rich environments (McNeal and Balistrieri, 1989). The tendency of selenium to undergo redox mediated precipitation is a dominant factor controlling the accumulation of Se in modern wetland sediments (i.e. Kesterson Reservoir) and in ancient wetland sediments (coals and lignites) (Horne, 1994; Ohlendorf and Santolo, 1994; Tewalt, 1986; Tokunaga et al., 1996).

In uranium areas of South Texas, selenium has been measured in soils and stream sediments at concentrations ranging from background to 16,000 ug/kg (Henry and Kapadia, 1980). In surface water, Se values from these areas have been found to range from background to 1 ppb (USDOE, 1995a). Selenium concentration in AML mine pit lakes were below the detection limit of 6 ug/L for all sites (Brandt and others, 1994).

**Vanadium:** Vanadium, (atomic number 23, atomic weight 50.94) is located in period 5 in the first row of the transition metals in the periodic table of the elements. As such, the zero-valent form of V has an electron configuration of (Ar)3d<sup>3</sup>4s<sup>2</sup> (Bello, 1988). Vanadium exists in the +3, +4 and +5 oxidations states (and a very rare +2) (Pais and Jones, 1997). Trivalent V is the most common form in the earth's crust (Kesler, 1994). Tetravalent V commonly occurs as vanadyl cation (VO<sup>2+</sup>), which acts like other simple divalent cations (Ca<sup>2+</sup>, Mn<sup>2+</sup>, Fe<sup>2+</sup>), etc. Pentavalent V usually found as the vanadate ion (VO<sub>3</sub><sup>-</sup> or H<sub>2</sub>VO<sub>4</sub><sup>-</sup>) the most stable environmental form (Hem, 1989). Vanadium concentrations in earth materials are strongly controlled by mineralogy. Even though V is more than twice as abundant (160 ppm) as copper (50 ppm) in the crust, vanadium occurs mostly as substituted ions in other minerals. Vanadium does not form many minerals, a fact that prevents ready extraction and use (Kesler, 1994). Instead, Vanadium abstraction, largely for use as a strengthening additive in steelmaking, is usually a byproduct of other extraction industries, including uranium mining (Goldberg et al., 1992).

Vanadium has been established as both an essential and a toxic element. Toxic effects include nervous system, liver, kidney and blood disruption (Nielsen, 1997) as well as reproductive and potential carcinogenic effects (Lu et al., 1998).

Vanadium oxidized by weathering processes (V<sup>5+</sup>) is highly soluble but will precipitate if the aqueous solution is evaporated or reduced - much like uranium (Kesler, 1994). Vanadium, like the other uranium-associated trace elements, is found in elevated concentrations in organic material including coal and petroleum (NAS, 1974) due to strong complexation and reduction reactions with humic substances (Lu et al., 1998). In the Colorado Plateau province prior to World War II, uranium deposits were initially mined as a source of vanadium. Only after the energy-rich nature of uranium was discovered in the late 1930s were the vanadium tailings reprocessed to extract the previously ignored uranium (Kesler, 1994).

Vanadate has been observed to occur in natural waters as an uncomplexed ion (Tanizaki et al., 1992). As an oxyanion, V tends to be highly soluble in oxidizing, alkaline waters, usually as the anionic forms VO<sub>4</sub><sup>3-</sup> and VO<sub>3</sub><sup>-</sup> (Pais and Jones, 1997). More reduced forms of vanadium have relatively low solubilities unless pH is below 4.0 (Hem, 1989).

Vanadium has not previously been recognized as a problem element in association with South Texas uranium mines (Brandt, 2000) and has not been routinely measured as a part of regulatory oversight by the RRC. Nevertheless, vanadium is suspected to be involved as a trace element emanating from uranium mining operations. Uranium vanadate minerals are described in early reports on the district (deVergie, 1958; Eargle et al., 1971). In

uranium areas of South Texas, vanadium has been measured in soils and stream sediments at concentrations ranging from background to 21,500 ug/kg. In surface water, V values from these areas have been found to range from background to 47 ppb (USDOE, 1995a). Vanadium concentrations were not determined in AML mine pit lakes (Brandt and others, 1994).

The environmental fate, toxicity and bioavailability of As, Mo Se and V in different media are controlled by a range of physical and chemical factors, including pH, redox potential (Eh), sediment mineralogy and grain size and the abundance and nature of sorbents and reactive particles such as clay minerals, organic matter and Fe and Mn hydrous oxides. In watershed systems, complex interactions among these factors result in a complicated yet predictable distribution of trace element species. On the basis of geochemical similarities, a conceptual model can be assembled to predict the behavior of U, As, Mo, Se and V within the drainages downgradient of the uranium mining areas. All of these elements are redox sensitive. All of these elements are solubilized by oxidizing conditions and are insolubilized by reducing conditions. All of these elements have a tendency to sorb to iron oxides. All of these elements tend to form strong complexes with organic matter. In the watersheds draining South Texas uranium mining areas, historic and present day accumulation of trace elements is most likely to concentrate these substances in locations along drainageways characterized by fine-grained siliciclastic deposition (silts and clays) and by the accumulation of organic carbon. These depositional characteristics will best be found in wetland environments.

#### **THE ENVIRONMENTAL IMPACT OF CONTAMINANTS RELEASED BY URANIUM MINING IN SOUTH TEXAS**

Uranium mining in South Texas resulted several documented adverse impacts to the local environment. Mining resulted in large-scale landscape destruction and the perturbation of pre-existing equilibria regarding the flux of uranium and associated contaminants across the coastal plain. This alteration resulted in liberation of contaminants that caused actual harm to the health of humans, livestock and the environment. In addition to actual adverse impacts, we contend that uranium mining also contributes to ongoing risk from concentration of liberated trace elements in areas that have not previously been investigated. One of our primary objectives for engaging in research on uranium mining is to determine the spatial distribution, the potential magnitude and the time function associated with contaminants released to the environment from uranium mining and milling.

Areas that are mineralized are also characterized by an elevated "background" concentration of these substances due to natural weathering and erosion (Alkhatib, 1998; Runnels, 1992; Henry and Kapadia, 1980). Mineralized areas are sometimes characterized by the accumulation of heavy metals and trace elements to levels of environmental concern from both natural and human agencies. Human intervention generally results in an accelerated rate of release of these substances to the environment. Accumulation of heavy metals and trace elements is most often observed in fine-grained sediments, in organic matter-rich deposits and in the biota (Carter, 1997; Lee, 1997; Horowitz, 1985).

Prior to open pit mining, there was a naturally elevated "background" signature of trace elements in the uranium mining areas of South Texas. Natural processes of chemical weathering, pedogenesis and erosion were slowly dismantling the near-surface roll front deposits resulting in a natural dispersion of materials from the ore. These processes, operating on geologic time scales, enhanced the concentration of uranium and associated trace elements within the soils, sediments, ground water and surface water downgradient of the mineralization. It is commonplace for

prospectors to use knowledge of these processes of dispersion to locate areas that are mineralized (Boyle, 1978). In the area where first discovered, the uranium deposits were essentially at the ground surface (Rotter, 2000). It is probable that several roll front deposits had been totally dismantled by natural processes over the last several hundred thousand years. If this is the case, then we would expect to find an extremely diluted signal recorded in sediments derived from the mineralized area. There has been no previous effort to establish the influence natural erosion has had upon the concentration of uranium and associated trace elements in coastal margin sediments.

In South Texas, most of the open pits were excavated below the local ground water table (Cossey, 1983). Ground water infiltration, captured rainwater and stormwater runoff necessitated pit dewatering in order for mining to occur (Galloway et al., 1979a; Texas Department of Agriculture, 1991; Ruttenber, 1984). Prior to passage of the Surface Mining Act, pit waters were usually discharged to nearby surface water drainages, releasing contaminants to the environment (Whadford, 1997; Manka, 1998). Dewatering effluent changed the hydrology of the receiving drainages, altering ephemeral streams into flowing watercourses (Wasserman et. al., 1996). Although not tested prior to regulation, it is reasonable to assume that dissolved constituents would have elevated concentrations in pit dewatering effluent. The composition of modern pit lakes among the inventory of unrestored AML mines may serve as an indication of the composition of dewatering effluent. The range of compositions is as follows: uranium (less than 1 to 4500 ug/L); arsenic (5 to 33 ug/L); molybdenum (10 to 650 ug/L) and, selenium (all less than 6 ug/L). Vanadium was not measured or reported in the RRC AML report (Brandt and others, 1994).

Dissolved contaminants discharged with dewatering effluent would be transported away from the mine vicinity. (This phenomenon has been observed in the Puerco River in New Mexico (Ruttenber, 1984; Graf, 1996)). Prior to 1975, many of the South Texas open pits were abandoned and allowed to fill with water. Near the first SWI mill, pits were converted into storage areas for tailings or processing wastes. Processing wastes introduced to unlined pits often acted to recharge ground water. In the Deweeseville area of western Karnes County, a processing mill delivered the acidic tailings slurries to former mine pits, resulting in two large plumes of low pH ground water (USDOE, 1995a; USDOE, 1998a) (Figure 12).

As this part of Texas is characterized by relatively dry conditions through most of the year, local officials sought to evaluate the water derived from pit lakes for future uses. Itin (1975) evaluated the potential for abandoned uranium pit lakes to serve as sources of drinking water, irrigation, livestock and wildlife water, fish and aquatic waterfowl habitat and recreational water. In spite of analyses for only the most general water quality indicators and radionuclides, Itin (1975) determined that the pit lakes would be unsuitable for any of these uses (Itin, 1975). In spite of these findings, local residents use existing pit lakes for recreational boating and fishing.

Molybdenum distributed to the nearby environment from tailings, mine dewatering effluent and processing slurries resulted in toxic effects upon livestock due to bioaccumulation in vegetation surrounding drainages. Consumption of Mo enriched vegetation by cattle grazing along streams near the uranium mines developed the disease molybdenosis (otherwise known as hypocuprosis, teart or peat scours) (Dollahite, 1972; USDOE, 1997a) (Figure 13) (Henry and Kapadia, 1980; Kaback, 1979). The Mo concentration in forage that results in disease, initially thought to be about 20 ppm (Barshad, 1948), is now known to be as low as 2 ppm (USEPA, 1979). Bioaccumulation of Mo in forage vegetation is accelerated in alkaline soils; such as occur in the South Texas uranium area,

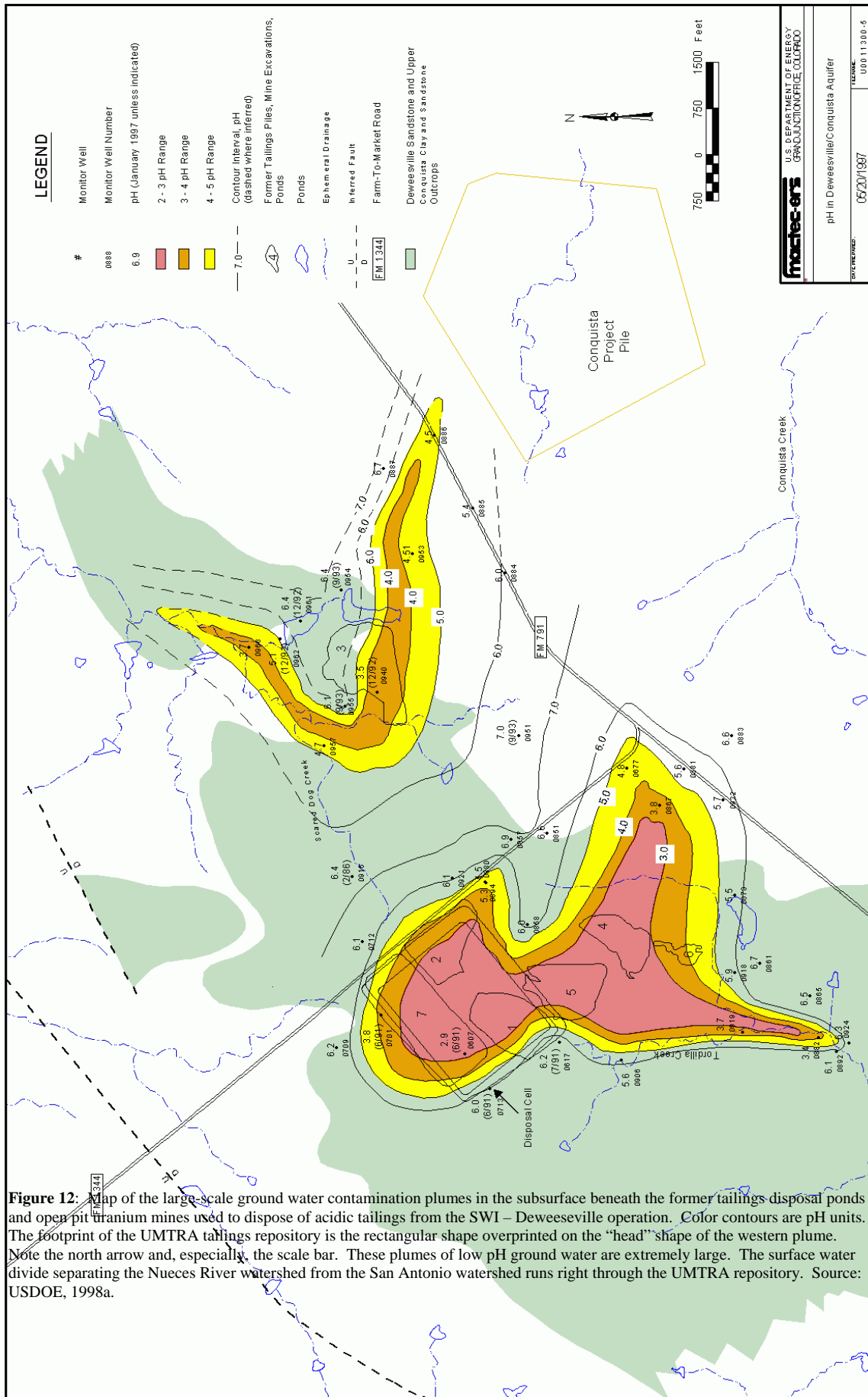
because of the increased dominance of molybdate ( $\text{MoO}_4^-$ ) in soil solution with increases in pH (Gupta, 1997).



**Figure 13:** Normal Black Angus calf (above) compared to calf born near SWI Deweeseville mill with achromotrichia due to molybdenosis. Note that the calf is not black. Sources: Above-unknown Internet; below-Leo Lyssy, Jr.

Molybdenosis is a potentially fatal disease in animals indicated by severe diarrhea, achromotrichia (a pronounced color change; generally a whitening), weight loss, retarded growth, loss of reproductive function, joint stiffness and shedding (Barshad, 1948). Molybdenosis, while most recognized as a disease of cattle, has also been observed in sheep, horses and humans (Erdman, 1990; Ladefoged, 1995) moose and deer (Frank, 1998). The disease is caused by molybdenum interference with metabolically required copper absorption in the gastrointestinal tract (Erdman, 1990). Copper deficiency is induced when forage has a Cu to Mo ratio less than 2. Excess Mo disturbing this ratio may be offset by increasing Cu in the diet (Johnson, 1997). Although the onset of molybdenosis in South Texas has been blamed on naturally occurring Mo (USDOE, 1995a), the association of the disease with uranium mining activities is unambiguous (Dollahite, 1972; Kaback, 1979; USDOE, 1997a).

In South Texas, molybdenosis was eradicated by supplying effected cattle with excess dietary Cu (USDOE, 1995). In spite of



**Figure 12:** Map of the large-scale ground water contamination plumes in the subsurface beneath the former tailings disposal ponds and open pit uranium mines used to dispose of acidic tailings from the SWI – Deweeseville operation. Color contours are pH units. The footprint of the UMTRA tailings repository is the rectangular shape overprinted on the “head” shape of the western plume. Note the north arrow and, especially, the scale bar. These plumes of low pH ground water are extremely large. The surface water divide separating the Nueces River watershed from the San Antonio watershed runs right through the UMTRA repository. Source: USDOE, 1998a.

the apparent disappearance of the disease, molybdenum is still found in soil and surface water at concentrations that have the potential to cause molybdenosis (Brandt and others, 1994; USDOE, 1995, our own data). It has been suggested that cattle grazing in the uranium-enriched areas of South Texas exist in a state of borderline molybdenosis (Hale, 1999). The potential impact of excess Mo on wildlife ruminants (deer) has not been satisfactorily investigated.

Molybdenosis identified in other areas has also developed (sometimes rapidly) as the direct result of uranium mining operations (USEPA, 1979; Erdman, 1990). In North Dakota, molybdenosis developed in cattle and sheep foraging on vegetation impacted by an ashing facility burning uraniumiferous Tertiary lignites. The ashing plant had been in operation for four years; the impacted livestock (cattle and sheep) had been exposed to contaminated forage for less than a year (Christianson, 1971).

Development of molybdenosis is a clear example of the linkage between geochemistry and toxicity. In Great Britain, the linkage between geology and disease was confirmed when the source of molybdenosis there was unequivocally correlated to dark colored shales and limestones of the lower Lias (Thornton, 1977). In Sweden, a mysterious disease among the Moose population was observed after the initiation of a liming program to counteract acidified lakes and wetlands. Diagnosed as molybdenosis, the disease resulted in numerous symptoms (including sudden heart failure) that proved fatal to over 1,500 moose since 1985 (Frank, 1998).

Development of molybdenosis near South Texas mines led to an investigation of the trace element geochemistry of the area surrounding uranium mines in Karnes County, Texas (Henry and Kapadia, 1980). Elevated concentrations of As, Mo and Se (U and V were not studied) were found to occur: in soils developed near uranium ore bodies; in fluvial sediments in drainages that received tailings runoff and pit-dewatering discharge from the mines; and, in some reclaimed areas (Henry and Kapadia, 1980). With the exceptions of the open pit mines themselves, the most elevated concentrations of trace elements were consistently recovered from stream sediments. In spite of this observation, the density of stream sample collection was insufficient to draw meaningful conclusions about the magnitude of trace metal flux within and through the drainage system. Because the objectives of the Henry and Kapadia (1980) study were to determine the probable causes of the toxic effects in livestock, there was no attempt to determine processes controlling environmental fate or bioavailability of As, Mo and Se.

South Texas uranium extraction has also been implicated as a cause of adverse health effects in humans living near mines (Au and others, 1995). In their study, Au and others (1995, 1998) used the technique known as molecular epidemiology to identify biomarkers indicative of increased exposure to radiation. This approach is relatively new, acting to determine potential exposures as a means of estimating risk of future disease rather than monitoring actual disease occurrences. The study was founded upon 24 individuals living within a ½-mile of a uranium mine and 24 control individuals. Controls were matched to study individuals based on age and gender. Environmental samples were collected from households and blood samples were collected for environmental and cytogenetic analyses. Analysis of environmental samples (surface soils, house dust and well water) revealed significantly higher concentrations of uranium compared to control households. From blood samples, 600 metaphase cells were analyzed for each individual. Cells from the exposed population had more chromosome aberrations than those from controls. After the cells were irradiated with gamma rays, cells from the exposed population revealed significantly more chromosomal aberrations than controls. This is described as indicating an abnormal DNA repair response (Au and others, 1998). The molecular epidemiology data is striking evidence that the legacy of uranium mining and milling may extend many years

beyond the phase of active mining. Determining the magnitude to which uranium-related contaminants have migrated away from the immediate area on mining and milling may suggest further investigation from molecular epidemiology.

From data acquired during the study of the UMTRA site, the USDOE established that stream transport of contaminants was evident in Tordilla and Scared Dog Creeks (tributaries of the Nueces and San Antonio Rivers respectively (USDOE, 1987; Texas Senate, 1989). While the Falls City UMTRA site was the subject of a large number of site specific investigations (Blount, 1992; Bryson, 1988; Ford, 1977; Ford, 1977; Haywood, 1980; Itin, 1975; Kallus, 1975; USDOE, 1983; USDOE, 1987; USDOE, 1990; USDOE, 1991a; USDOE, 1991b; USDOE, 1992a; USDOE, 1992b; USDOE, 1992c; USDOE, 1994a; USDOE, 1994b; USDOE, 1995a; USDOE, 1995b; USDOE, 1996a; USDOE, 1996b; USDOE, 1996c; USDOE, 1997a; USDOE, 1997b; USDOE, 1997c; USDOE, 1998a; USDOE, 1998b) these investigations were limited to a two mile radius from the UMTRA site (with most of the investigation activity confined to a one mile radius). The UMTRA enabling legislation also required remediation of "vicinity" properties; this directive was not interpreted to include off-site migration of trace elements (U.S. House of Representatives, 1996).

In spite of many studies conducted in South Texas evaluating environmental contamination from uranium mining and milling, no studies have investigated the environmental fate of trace elements in watersheds beyond the immediate vicinity of the open pits. In order to better define the potential distribution of uranium mining-related trace elements within the watershed, data is being assembled on a GIS to describe the spatial distribution of important features. This article presents the historical and geochemical background upon which the research effort is based. At the 2000 South Texas Environmental Conference we will present preliminary data from the on-going research. The results of our research will fill a gap in the knowledge base regarding the potential for continued release of contaminants to and through watersheds that have witnessed uranium mining and milling. Filling this gap will promote a better understanding of mechanisms controlling trace element flux and will fortify resource management strategies to protect the vast environmental and economic importance of Texas Gulf Coast freshwater and estuarine systems.

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Ron Parker is a geologist working on his doctoral degree at Texas A&M University in College Station. His research targets the environmental geochemistry and watershed-scale dispersion of trace elements liberated from South Texas Uranium Mines. He is

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Bruce Herbert is an Associate Professor of Geology in the Department of Geology and Geophysics at Texas A & M University. Dr. Herbert has been at Texas A & M since 1992 and specializes in environmental geochemistry. His research interests involve the molecular-scale mechanisms that control contaminant distributions in environmental media. He has research interest in both inorganic and organic contaminants. Dr. Herbert has a Ph.D. from the University of California – Riverside where he conducted research on water-soluble organic carbon under Dr. Garrison Sposito and Dr. Paul Bertsch. He has a B. A. in Chemistry from Colgate University. Prior to accepting a position at Texas A & M, Dr. Herbert worked at the Savannah River Ecology Laboratory.

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