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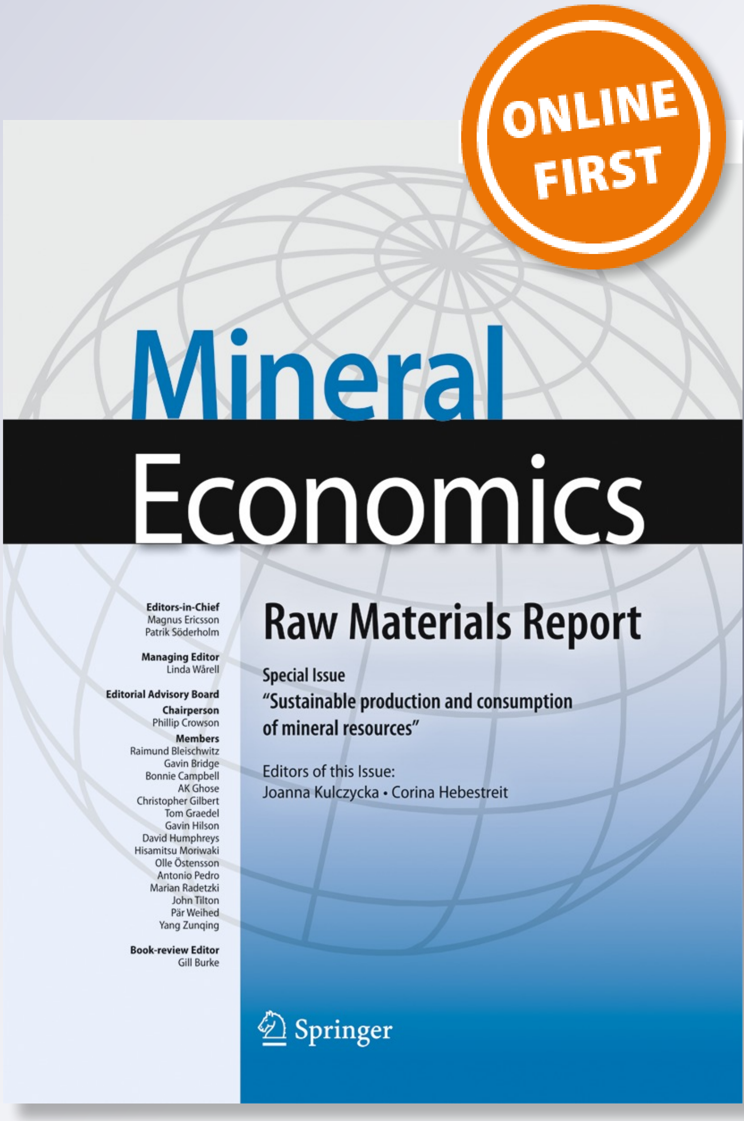

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ORIGINAL PAPER

Potential recovery of aluminum, titanium, lead, and zinc from tailings in the abandoned Picher mining district of Oklahoma

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Abstract The abandoned Picher mining district in northeastern Oklahoma, part of the Tri-State mining district, was the largest source of sulfide ores of lead and zinc in the U.S. A. in the early twentieth century. After abandonment in the 1960s, numerous environmental problems caused by the abandoned mines and large tailings piles affected the district and surrounding areas with contamination of water, soils, vegetation, wildlife, and humans with lead and other metals. Current (2011) cleanup efforts in the district include separation of coarse and fine tailings particles, sales of the coarse particles (which consist mostly of dolomite and chert) for use as aggregate in asphalt, and burial of metals-rich fine tailings below ground to reduce exposure to and transport of toxic metals. Analysis in this paper indicates that reprocessing these tailings to extract remaining aluminum, titanium, lead, and zinc may be feasible. The value of lead and zinc in the tailings of this abandoned mining district probably are not sufficient to justify recovery of metals from the tailings at current market prices, but potential recovery of aluminum and titanium from these tailings may provide sufficient returns to justify the costs associated with reprocessing the fine tailings remaining in this district.

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Introduction

Mining has played a vital role in the discovery, settlement, and development of the U.S.A. (Robertson 2006). Due to more than two centuries of mining, the U.S.A. has numerous abandoned mining districts having tens to hundreds of square kilometers of land with extensive degradation in the quality, diversity, health, and productivity of soils, vegetation, wildlife, and water. The Picher mining district, located in northern Ottawa County, Oklahoma (Fig. 1), was part of the Tri-State mining district, a 3,080-km^2 area in southwest Missouri, southeast Kansas, and northeast Oklahoma that was the most productive site of mining for sulfide ores of lead and zinc in the U.S.A. from the late 1830s to the 1970s (Gibson 1972; Gibson 1982; Robertson 2006). The Picher mining district, included in the initial National Priority List of Superfund sites as the Tar Creek Superfund site by the U. S. Environmental Protection Agency in 1983, is plagued by some of the most severe or widespread environmental degradation of any of the tens of thousands of abandoned mining sites in the U.S.A. (Ferderer 1996; State of Oklahoma 2000; Robertson 2006). The district is on the northwest side of the Ozark Uplift and is underlain by 30 or more meters of shales of Pennsylvanian age and a sequence of more than 300 m of carbonate rocks of Cambrian, Ordovician, and Mississippian age (McKnight and Fischer 1979; Marcher and Bingham, 1971). The area is drained by numerous creeks and the Neosho and Spring Rivers (Gibson 1972). Topographic relief in the district is gently rolling, comprised of small hills and incised stream valleys (Gibson

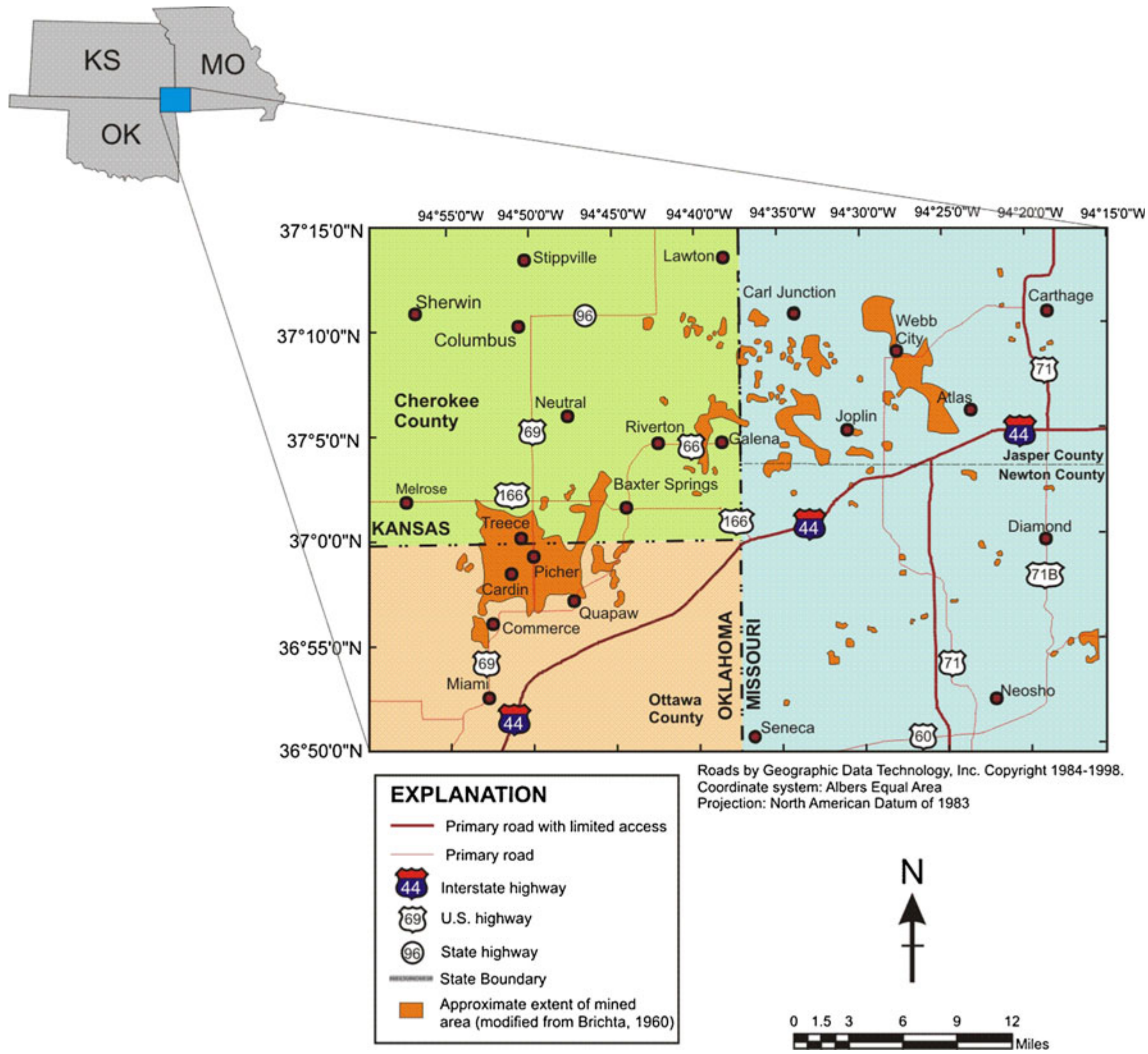

Fig. 1 Location of Tri-State mining district and the Picher mining district in Oklahoma

1972, 1982). Hills in the area are heavily wooded with mixed stands of white oak, blackjack oak, hickory, and sycamore trees, with the mild, temperate continental climate of the district facilitating year-round mining (Gibson 1982).

Mining history

Mining of these Mississippi Valley type deposits started in cherty limestones and dolomites in the Missouri portion of the Tri-State mining district, gradually progressing westward in the late 1800s and early 1900s to the Kansas and Oklahoma parts of the district. Initially, mining concentrated on recovery and smelting of the lead–sulfide (PbS) mineral galena due to its ready identification in the field, easy separation from the host rock due to its great density, low temperatures at which it can be smelted, high purity of the ore, and demand for lead for bullets, paints, pipes, and many other uses (Gibson 1982). When zinc smelters opened in 1872 in St. Louis, Missouri and in Illinois, the zinc sulfide mineral sphalerite, which had been discarded as waste rock, became a profitable commodity. As the price of zinc rose from $3 per ton in 1872 to $27 per ton in 1888, zinc mining surpassed lead mining in the district and mines increased in depth from the water table, which occurred about 15 m below land surface, to more than 90 m below land surface, with zinc ore being more abundant below the water table (Gibson 1972). Miners followed veins and cavities of the richest ore, commonly cutting stopes 23 m or more in height (Fig. 2). After the larger mining companies ceased operations in the 1950s, "gougers" (small private miners) continued to mine small ore deposits through the 1960s (Fig. 3), sometimes removing support pillars in the process.

Post-mining period

When mining stopped ca. 1970, as much as 165–300 million tons of mine tailings, some of which had been reprocessed

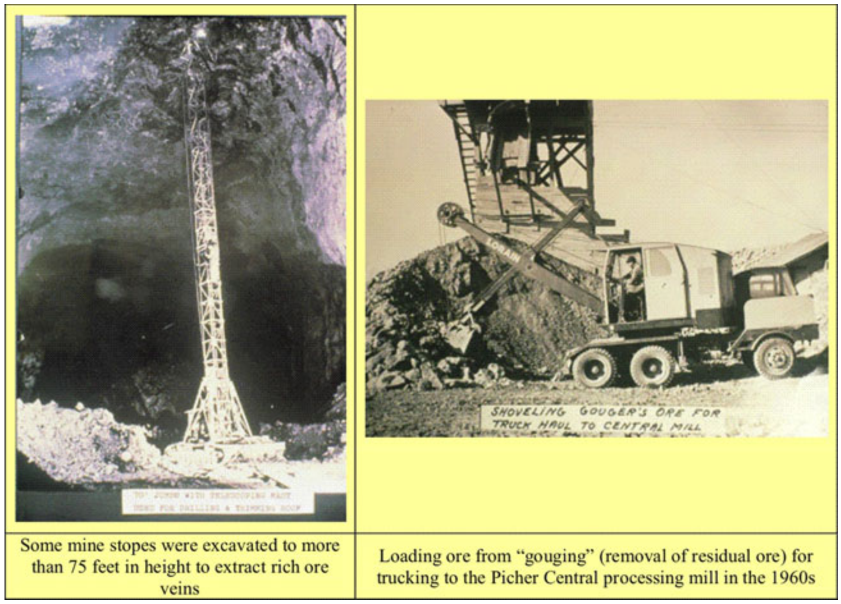

Fig. 2 Photographs of mining in the Picher mining district (photographs from F.J. Cuddeback)

one or more times to recover metals, remained in the Picher mining district (State of Oklahoma 2000; Netzeband 1936). By 2000, the U.S. Army Corps of Engineers estimated that 60 million cubic yards or 75 million tons of mine tailings remained in the district (State of Oklahoma 2000). By 2008, with continued removal of tailings, estimates of tailings remaining in the district were about 24 million cubic meters or 47 million tons (U.S. Environmental Protection Agency 2008). Mine tailings in the district have primarily been considered to be a nuisance that potentially exposes humans, wildlife, and the surrounding ecosystem to hazardous metals such as cadmium and lead and to silica dust, a potential cause of the lung disease silicosis.

From the time of mining to the present, boulders and finer tailings remaining in the mining district (Fig. 3) have been progressively mined, primarily for use as low-cost ($0.50–1.50 per cubic yard (approximately 1.4 metric tons)) aggregate for use in hundreds of miles of unpaved and asphalt-paved roads, rip-rap to prevent streambank erosion, railroad track ballast, and in concrete (Murdock 1929; Cobban 1934; Fenimore 1934; Langerhans 1934; Smith 1934; Netzeband 1937). Additional uses of smaller size fractions of mine tailings from the district have included sand for coating stucco, blasting sand, rock sawing sand, filler in composition roofing, gravel in tar/gravel flat roofs, and sand spread for traction on icy roadways (Netzeband 1937). Those finer tailing fractions, which were widely used across the region, are more likely to have contained greater concentrations of metals, including relatively toxic metals such as lead and cadmium.

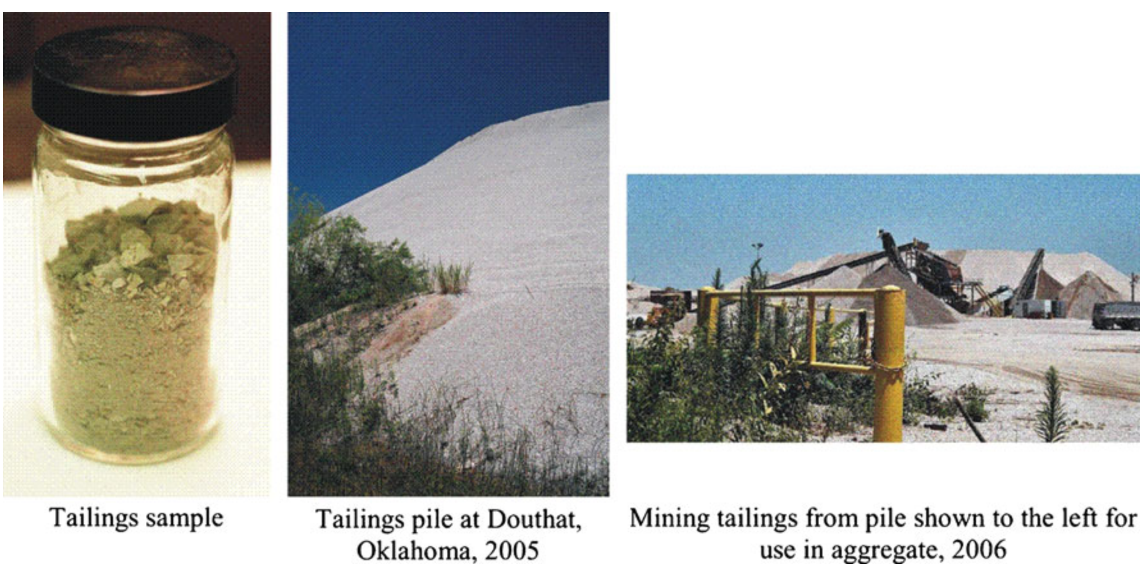

Fig. 3 Photographs of tailings and tailings excavations taken by the authors in the Picher mining district

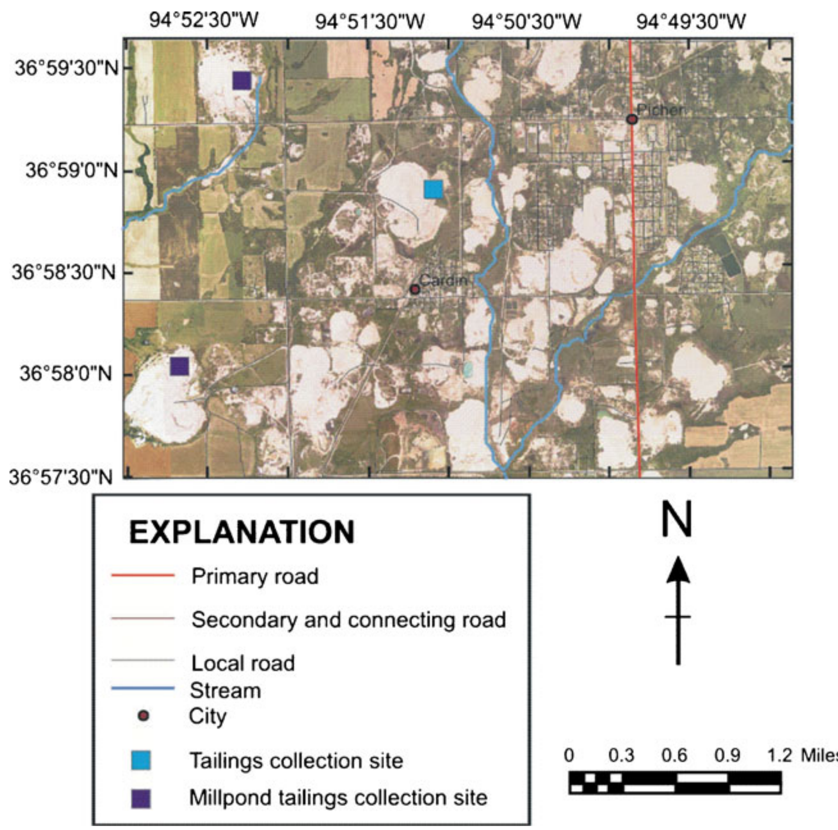

Fig. 4 Map of selected collection sites for tailings analyzed by the University of Oklahoma from 2004 to 2007

By the late 1940s, as mining was becoming economically marginal, estimated reserves of 2.4 million tons of zinc concentrates and more than 300,000 tons of lead concentrates (in the form of galena and sphalerite) were believed to remain underground in the Tri-State mining district (Tri-State Ore Producers' Association 1946). As noted by Bell and Donnelly (2006), mineral deposits abandoned when they become uneconomic to mine may be reworked in the future when mining technology and demand makes their extraction worthwhile again. This paper was written to investigate that possibility of reprocessing the easily accessible mine tailings remaining on the land surface in this district.

Analysis of mine tailings

In 2004, students and faculty from the School of Civil Engineering and Environmental Science at the University of Oklahoma, in cooperation with the U.S. Environmental Protection Agency and the Oklahoma Department of Environmental Quality, conducted field experiments indicating that incorporation of unsorted tailings into asphalt used in road pavement was an effective way to reduce runoff of metals from unconsolidated tailings-covered roads and to encapsulate hazardous metals primarily associated with fine particles in the tailings. A disposal method that has been tested for the metals-rich fine tailing particles is re-injection into mine workings to remove tailings from the land surface (Meyer 2005; U.S. Environmental Protection Agency 2008). Possible problems with re-injecting tailings into the mine workings include potential influx of large amounts of easily transported metals-rich fine particles in local ground water and connected streams and the preclusion of other potential uses of the tailings.

As part of studies to determine potential uses of mine tailings and cycling of metals in the environment in the mining district, analyses were made of concentrations of lead, cadmium, zinc, and other metals in unsieved and sieved tailings and of fine tailings remaining in flotation ponds by students and

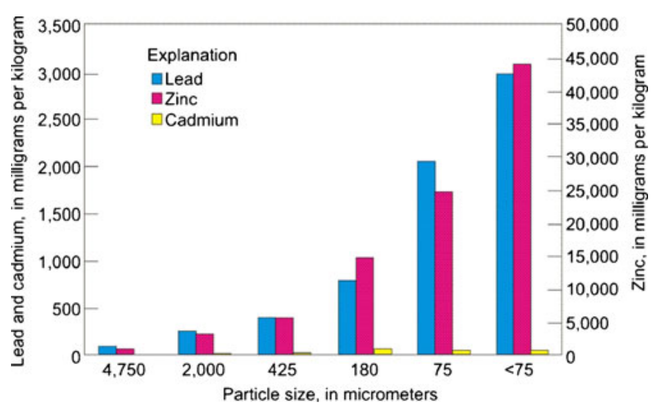

Fig. 5 Concentrations of lead, cadmium, and zinc in a mine tailings sample collected from the Kenoyer North tailings pile in Ottawa County, Oklahoma

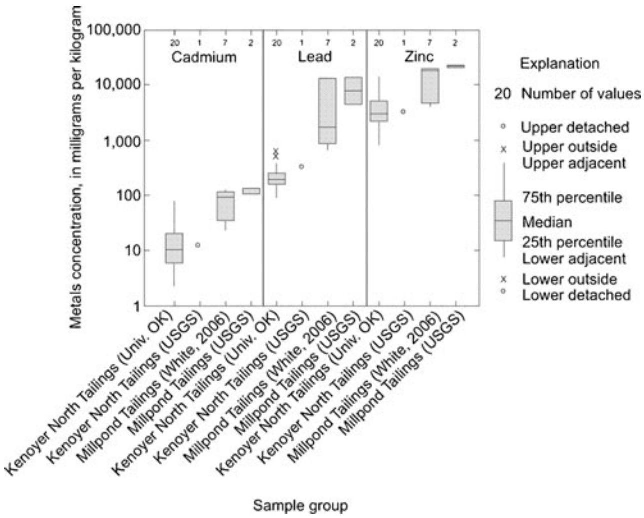

Fig. 6 Boxplots of concentrations of cadmium, lead, and zinc in tailings samples analyzed by the University of Oklahoma (OU) (LaBar and Nairn 2007), by a U.S. Geological Survey Laboratory in 2005–2006, and in millpond tailings analyzed in White (2006)

faculty from the University of Oklahoma (White 2006; LaBar 2007). Analyses of unsieved tailings collected from the Kenoyer North pile (Fig. 4) from 2005 to 2006 indicated that concentrations of lead in those tailings exceeded a 500-mg/kg cleanup standard for residential parts of the mining district specified by the U.S. Environmental Protection Agency. Metals concentrations have typically been reported to be more concentrated in fine particles (<2-mm diameter) in local mine tailings, with larger particles being dominated by chert and dolomite (Fig. 5).

To determine metals that might be present in mine tailings in the district, one tailings sample from the Kenoyer North pile (sieved to remove particles greater than 2 mm in diameter) and two fine tailings samples from a millpond southwest of Cardin, OK (Fig. 4) were digested with nitric acid, heated by microwave (U.S. Environmental Protection Agency 1994), and analyzed by inductively coupled plasma mass spectroscopy (ICP-MS) at the USGS Mineral Research Laboratory in Lakewood, CO. Metals analyzed by ICP-MS included: silver, aluminum, arsenic, barium, beryllium, bismuth, calcium, cadmium, cerium, cobalt, chromium, cesium, copper, iron, gallium, germanium, potassium, lanthanum, lithium, magnesium, manganese, molybdenum, sodium, niobium, nickel, phosphorus, lead, rubidium, tin, scandium, thorium, titanium, thallium, uranium, vanadium, yttrium, and zinc. Detection limits for those metals ranged from 0.003 to 100 mg/kg, with most metals having detection limits less than 1 mg/kg.

As shown in Fig. 6, the three samples analyzed for this paper had similar concentrations of cadmium, lead, and zinc as other tailings samples collected in the mining district by the University of Oklahoma students since 2004. Concentrations of these metals are similar to those described in Oklahoma Department of Environmental Quality (2003) for unsorted tailings and fine particles sieved from tailings from two other tailings piles in the mining district.

Analysis of metals values

To evaluate potential economic values of metals in tailings and tailings fines, current or recent metals prices were obtained from Kitco and MetalPrices.com in October 2012 (Kitco 2012a, b, c, d). As shown in Tables 1 and 2 and Fig. 7, tailings in the Picher mining district contain values of aluminum, titanium, lead, and zinc substantially exceeding the $1.00–2.00 per metric ton that has been paid for unwashed tailings used in various types of aggregate.

Concentrations of lead (in the millpond samples) and zinc were notably greater than those reported by Kabata-Pendias and Pendias (1984) for clay and clay loam soils in the United States (Table 3). Aluminum and titanium may be useful by-products of the smelting process for lead and zinc. Values of these four metals ranging from about $12 to more than $117 per metric ton in fine particles from the Kenoyer North tailings pile and the millpond tailing samples (Tables 1 and 2, Fig. 7) far exceed the current market value for unwashed tailings of $1.00 to $2.00 per ton. Such value, with potential for increased value with time, indicates that there may be better

Table 1 Concentrations, recent prices, and estimated value of lead and zinc per metric ton of tailings from the Kenoyer North pile near Cardin, OK

Metal	Metals price, in dollars/kg	Sieved concentration, in mg/kg	Mass of metal per tonne of tailings, in kg	Metals value per tonne, in dollars	Unsieved concentration, in mg/kg	Mass of metal tonne, in kg	Metals value per tonne, in dollars
Aluminum[a]	$1.87	4,100	4.10	$7.67	1,700	1.70	$3.18
Titanium[b]	$10.50	139	0.139	$1.46	57.7	0.06	$0.63
Lead[a]	$2.01	890	0.890	$1.80	369	0.369	$0.74
Zinc[a]	$1.80	8,750	8.75	$17.13	3,630	3.63	$7.12
Theoretical value of selected metals per metric ton				$28.06			$12.29

[a] Kitco (2012a)

[b] MetalPrices.com (2012)

Table 2 Concentrations, recent prices, and estimated value of lead and zinc per metric ton of tailings from two millpond tailings samples collected near Cardin, OK

Metal	Metals price, in dollars/kg	Mean metal concentration, in mg/kg	Mass of metal per metric ton, in kg	Metals value per metric ton, in dollars
Aluminum[a]	$1.87	20,300	20.3	$37.96
Titanium[b]	$10.50	1,690	1.69	$17.74
Lead[a]	$2.01	9,180	9.18	$18.54
Zinc[a]	$1.80	22,300	22.3	$43.71
Selected metals value per metric ton				$117.95

[a] Kitco (2012a)

[b] MetalPrices.com (2012)

use for these metals-rich fine particles than disposal in underground mine workings or incorporation into asphalt and other aggregates.

Metals as strategic resources

Although the majority of lead and zinc consumed in the U.S.A. already comes from the U.S.A., most of titanium and much of the aluminum consumed in the U.S.A. comes from other Nations (Fig. 8). The U.S.A. would benefit from additional supplies of those metals, particularly considering the availability for reprocessing of millions of tons of these metals at the land surface near major transportation routes.

Gradual recovery of the prices of lead and zinc with economic recovery (Fig. 9), possible future events such for decreased imports due to increasing transportation costs and future conflicts, and continued improvements in ore processing technologies indicate potential for reprocessing tailings in the Picher mining district for remaining lead and zinc contents. Because the metals-rich tailing fines are on the land surface and finely ground, at least some of the costs typically associated with mining of metals ores would not be incurred in reprocessing these materials. The proximity of the mining district to a major interstate highway, rail lines, and the Kerr-McClellan-Arkansas River system connecting to nearby major industrial centers such as St. Louis, Missouri; Kansas City, Kansas; and Tulsa, Oklahoma and more distant points of use can reduce transportation costs of these materials to potential points of reprocessing or end users.

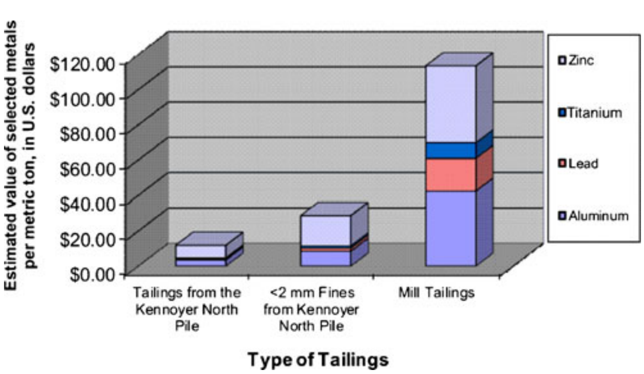

Fig. 7 Estimated value of lead and zinc per metric ton of mine tailings, sieved tailings fines, and millpond tailings from samples collected near Cardin, OK, in 2005

Table 3 Ranges of dry-weight concentrations of lead and zinc in clay and clay–loamy soils, or argillaceous sediments in North America (Kabata-Pendias and Pendias 1984)

Metal	Range of concentrations, in milligrams per kilogram
Aluminum	72,000–100,000
Titanium	1,470–14,700
Lead	10.0–70.0
Zinc	20.0–220

Conserving several million tons of metals-rich fine tailings in covered, monitored landfills may be useful as a part of a national strategy to maximize use of readily available base-metal resources in the U.S.A. Obtaining tens of dollars for metals content per ton of tailings, rather than a dollar or less per metric ton also seems likely to benefit the local economy, which is depressed as is typical for abandoned mining districts. Perhaps treating these metals-rich materials as liabilities, rather than as assets of substantial value, has been short-sighted.

Recovery of selected metals

The economic viability of recovery of metals from tailings in this district is affected by several factors including: plant cost, utilities cost, metals concentrations in tailings, refined metals spot prices, and transportation costs. The cost of a processing plant capable of processing 80,000 tons/year is estimated to range from 16 to 20 million US dollars. Given an approximate remaining amount of 8 million tonnes of tailings with 4.5 % zinc content, the amount of recovered zinc should be 3,600 tonnes per year. Given that a concentrate produced by the Waelz process (Ruetten, 2010) would contain 50 % zinc content, the amount of concentrate produced would be 7,200 tonnes per year, with the produced material being 0.09 tonnes per fine tailing tonne processed.

Given those estimates, worst case and best case scenarios of plant receipts can be based on a possible range of tailing concentrated prices. If the cost of concentrate production, in a existing plant, is 165 USD/tonne of fine mine tailings, and the price of Waelz concentrate is about 2,800 USD/tonne, it is estimated that earnings will be about 82 USD/tonne of fine mine tailings. Annually, such a plant is estimated to

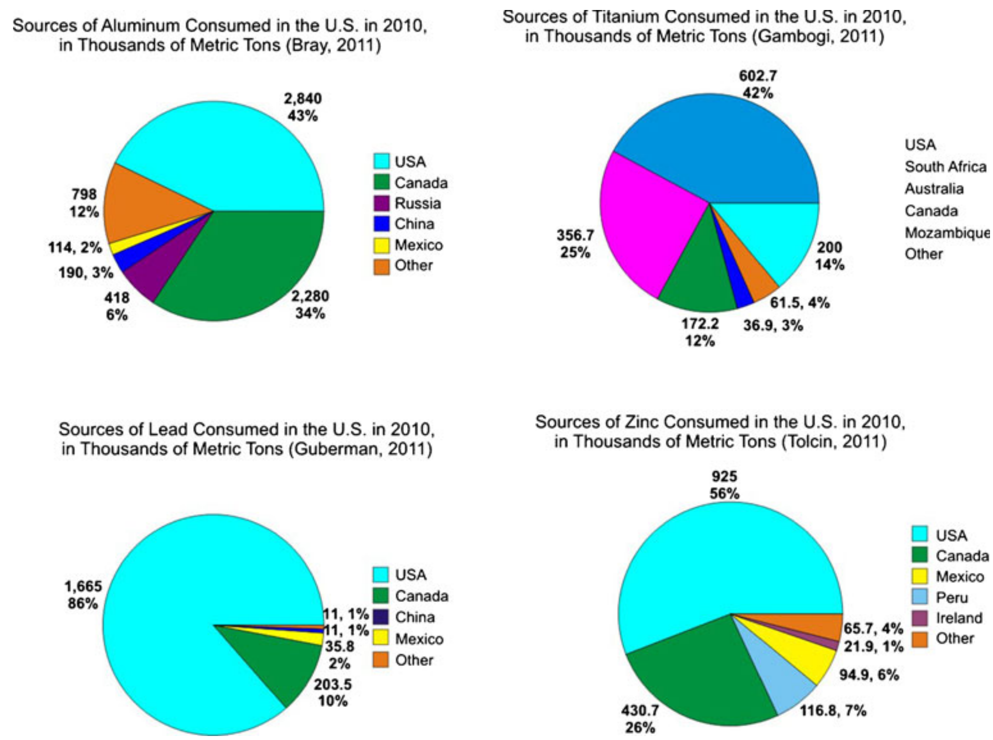

Fig. 8 Sources of lead and zinc consumed in the U.S.A. in 2006, in thousands of metric tons

earn 0.59 MUSD. In a best case scenario, the earnings of such a plant could be as much 2.1 MUSD per year.

If a bigger plant is used, the cost will increase less than linearly, and the earnings will be linear with production. That is, if the plant capacity were to be increased to 160,000 tons of fine mine tailings, the earnings would range from more than 1.5 MUSD to more than 5.0 MUSD.

Similar computations can be made with having to purchase coarse tailings for 1 USD/tonne. At a processing rate 400,000 tonnes per year, plant earnings would be 0.4 MUSD per year. If the production were doubled, the earnings would be more than 1 MUSD per year. Taken as a whole, potential annual earnings from the process could be between 1.0 and 2.5 MUSD per year and if plant production was again doubled, to between 2.5 and 6.5 MUSD per year.

Given these assumptions, a tailings reprocessing plant at the abandoned Picher mining district is likely to produce profits over the range of foreseeable zinc concentrate prices, with the initial investment in plant construction likely to be from 3 to 5 years, which may not be feasible or attractive to potential investors. However, if the value of titanium in the ores is considered, the feasibility or profitability of such a plant is likely to be improved. Titanium in the fine mine tailings comprises about 70 % of the total titanium in the tailings. About 8,176 tonnes of Ti could be recoverable, being concentrated in the Waelz process metallic material as titanium oxide. Another potentially recoverable metal is aluminum, about 165 tonnes of which would be recoverable using the same process. Given the value of these metals, the price of this processed ore would be about 50–100 USD/tonne. The Ti and Al in the tailings, in addition to the well-known Pb and Zn content, substantially improve the economic feasibility of recovery of metals from these tailings. The Ti and Al could be extracted with an electrolytic process. The final decision will depend on a test, because the solubility of the Ti and Al compound will be the key parameters for determining recoverability of those metals.

Given these limitations, the only way to recover the Zn, Pb, and Cd economically and to solve the environmental problem is to use an existing Waelz process plant, which will eliminate the need to recover the initial investment in plant construction. Further testing of the metals content and recoverability with separation and smelting tests, however, may indicate higher concentrations of metals that may increase economic feasibility of metals recovery from these tailings. If results of further testing support metals recovery from these tailings, then testing at an existing plant using the Waelz process would be useful.

Another strategy, in which the fine tailings containing 4. 5 % zinc are sold as ore for 30–50 US dollars per tonne, also may be feasible. In such a scenario, the coarse tailings might be sold for 0.4 million US dollars and the fine tailings might be sold for 2.4–4.0 million US dollars. Constructing a screening plant to process 80,000 tons/year would cost about 5 million US dollars, but there are several such plants already operating in the district, so such additional investment may not be unnecessary. Operating costs for separation would be 30–50 US dollars per tonne or 2.4 to 4.4 million US dollars per year. Selling the fine tailings to an outside processing plant would have the best return period, but that strategy might not be as profitable as operating a Waelz process plant in or near this mining district.

Summary

The abandoned Picher mining district, a major source of mining for ores of lead and zinc during the first half of the twentieth century, contains tens of millions of metric tons of mine tailings of varying metals content. From the 1920s through the early twenty-first century, tens of millions of tons of those tailings have been removed for relatively low-value uses as aggregates or re-injection into mine workings. Despite being reprocessed one or more times, tailings in the mining district contain substantial amounts of metals, ranging in concentrations up to tens of thousands of milligrams per kilogram (parts per million).

The value of lead and zinc in the tailings of this abandoned mining district probably are not sufficient to justify recovery of metals from the tailings at current market prices. However, if recovery of aluminum and titanium are determined to be feasible, then metals may be profitably recovered from these tailings, perhaps producing several million dollars per year.

Current metals values and potential future increases in prices support the conclusion that current sales of mine tailings at prices less than a few dollars per ton or disposal of metals-rich fines into underground mine workings are unlikely means of obtaining full value for this resource. Such practices, particularly underground disposal, also may have unanticipated environmental consequences, distributing these potentially toxic metals to the environment in unforeseen ways. Obtaining greater value for the residual metals in the tailings would not only reduce the national trade deficit, but would benefit local communities, which have lost thousands of residents since metals mining ceased, and which remain economically depressed relative to national median income, housing values, and infrastructure (Robertson 2006; Bureau of the Census 2011).

Extraction of aluminum, titanium, lead, and zinc in fine tailings particles and mill tailings in the Picher mining district may be feasible at current prices or at increased future prices. Developments in mineral-extraction technologies also may increase the likelihood that substantial further value could be obtained for materials currently considered to be wastes. Reserving some portion of these metals-rich fine tailings in contained landfills may be prudent as part of a National reserve of easily accessible base metals.

Fig. 9 Lead and zinc prices from 2002 to 2011, from Kitco (2012a, b, c, d) and MetalPrices.com (2012)

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