PASSIVE TREATMENT OPPORTUNITIES IN A DRASTICALLY DISTURBED WATERSHED: REVERSING THE IRREVERSIBLE?[1]

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Abstract: Like many large abandoned mining areas, the Tri-State Mining District of the central US is plagued by numerous environmental problems, including contaminated soils, tailings, waste rock, and water, with subsequent detrimental impacts to human and ecological health. In one of the major basins of the district, the Tar Creek watershed, impacts to surface waters were once deemed to be due to "irreversible man-made damages", resulting in minimal effort to address environmental risk from these legacy mine waters over the past 30 years. However, recent watershed-scale environmental monitoring studies, along with completion and evaluation of a full-scale passive treatment demonstration project, indicate that this may no longer be the case. Evaluation of artesian discharges indicates significant evolution of mine pool water quality (e.g., decreased metal concentrations, increased pH and alkalinity) since first flush. In-stream water quality, although still not meeting designated beneficial uses, has also changed. Substantial surface water degradation due to waste rock and tailings runoff and leachate is still problematic, but large-scale land reclamation (including wholesale removal and underground injection) is ongoing. Conceptual designs for watershed-scale passive treatment implementation have been developed and indicate that both artesian mine discharges and remaining runoff/leachate waters may be treatable in a cost-effective and sustainable manner. Recent voluntary relocation efforts, resulting in the dissolution of historic mining communities, provide opportunities for watershed-scale remediation and restoration. However, long-term operation and maintenance obligations for the proposed passive systems, incorporating the cultural and social needs of Native American tribes, requires sustained commitment of technical and financial resources. Promising initial results, coupled with results from other ongoing remediation efforts, warrant further exploration and reevaluation of previous administrative decisions.

Additional Key Words: acid mine drainage, constructed wetlands, basin remediation and restoration

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[1] Paper was presented at the 2011 National Meeting of the American Society of Mining and Reclamation, Bismarck, ND ***Reclamation: Sciences Leading to Success*** June 11 - 16, 2011. R.I. Barnhisel (Ed.) Published by ASMR, 3134 Montavesta Rd., Lexington, KY 40502.

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Proceedings America Society of Mining and Reclamation, 2011 pp 450-468
DOI: 10.21000/JASMR11010450
http://dx.doi.org/10.21000/JASMR11010450

Introduction

Like many abandoned hard-rock mining areas, the Tri-State Lead-Zinc Mining District of the central United States presents considerable environmental challenges, including contaminated surface and ground waters, waste piles, fines impoundments and collapse features (e.g., Nairn et al., 2010a; EPA, 2010). Straddling the boundaries of three states (Oklahoma, Kansas and Missouri) and two U.S. Environmental Protection Agency (EPA) regions (Regions 6 and 7), the jurisdictional complexities of the mining district are often cumbersome and convoluted, especially considering the presence of significant Native American land holdings in Oklahoma (e.g., USFWS, 2009). The district includes four U.S. EPA Comprehensive Environmental Response Compensation and Liability Act (CERCLA or Superfund) sites: Oronogo-Duenweg and Newton County Mines (Missouri), Cherokee County (Kansas) and Tar Creek (Oklahoma).

The Tar Creek Superfund Site includes the former Picher Field and the Tar, Lytle, Beaver, and Elm Creek watersheds (Fig. 1). The upper Tar Creek watershed was the site of intensive underground room and pillar mining for nearly a century and is characterized by large waste or "chat" piles, extensive un-vegetated fines impoundments, metals-contaminated artesian mine water discharges and collapse features (Fig. 2). The Tar Creek site was placed on the National Priorities List in 1983 and five Operable Units (OUs) have been established to facilitate clean up (EPA, 2010).

The Record of Decision (ROD) for OU 1 Surface and Ground Waters was issued in 1984. Subsequently, the Oklahoma Water Resources Board (OWRB) concluded that impacts to surface waters were due to "*irreversible man-made damages*" resulting from past mining operations (EPA, 1994, 2005, 2010). In 1985, OWRB lowered the designated use for surface waters from "Warm Water Aquatic Community" to "Habitat Limited Fishery" and "Secondary Recreation Water Body". The Oklahoma Water Quality Standards (OWQS, 2010) allows use re-designation when "*human-caused conditions of pollution prevent the attainment of the use and cannot be remedied.*"

Under the OU 1 ROD, U.S EPA utilized a fund-balancing waiver, based on the declaration that costs were prohibitively high to address surface water contamination. In essence, actions to address environmental risks were not warranted as the use re-designation was protective of human health and estimated costs to address other risks would drain the Superfund and risk the

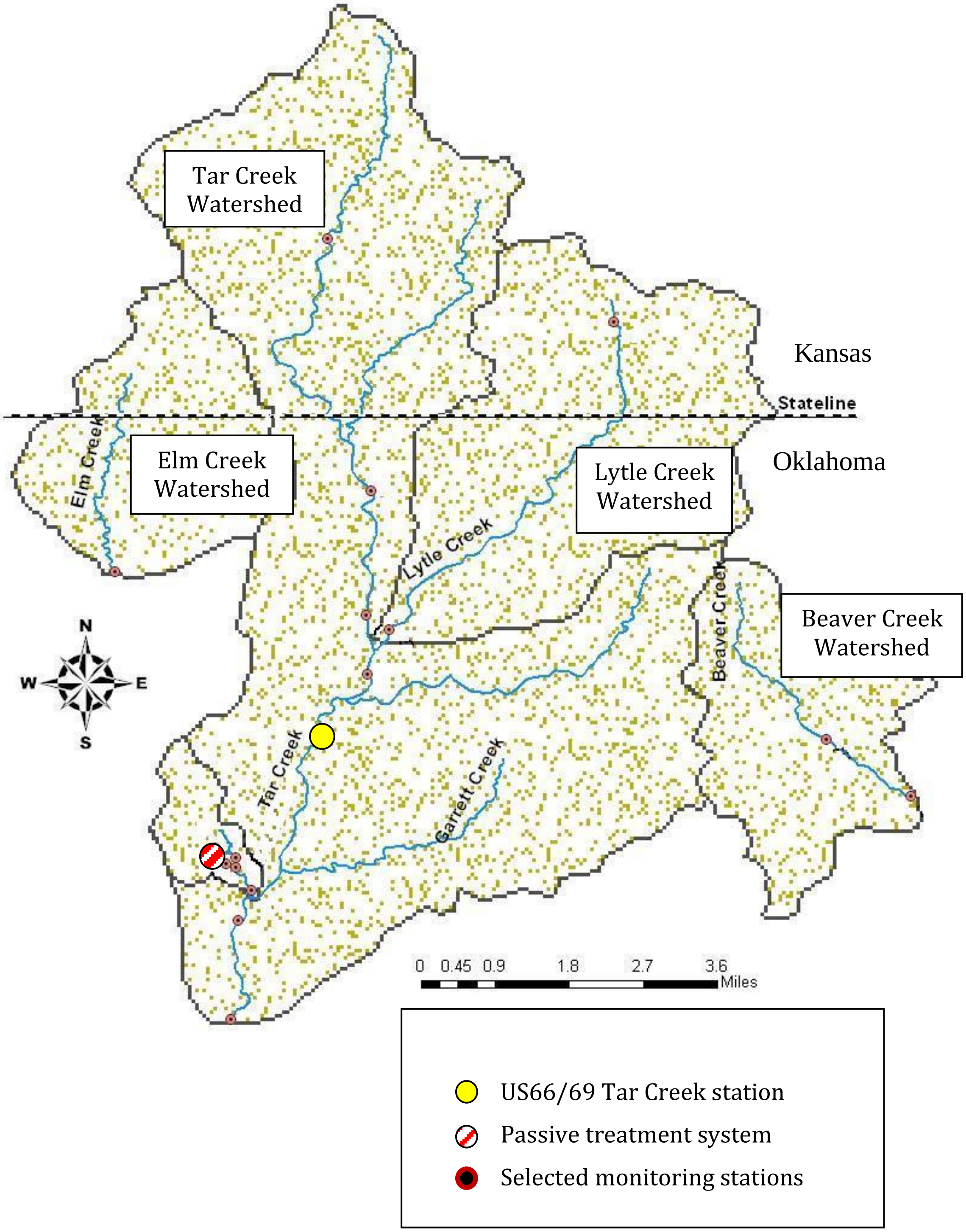

Figure 1. Major watersheds of the Tar Creek Superfund Site, Ottawa County, OK, illustrating selected sampling and study locations.

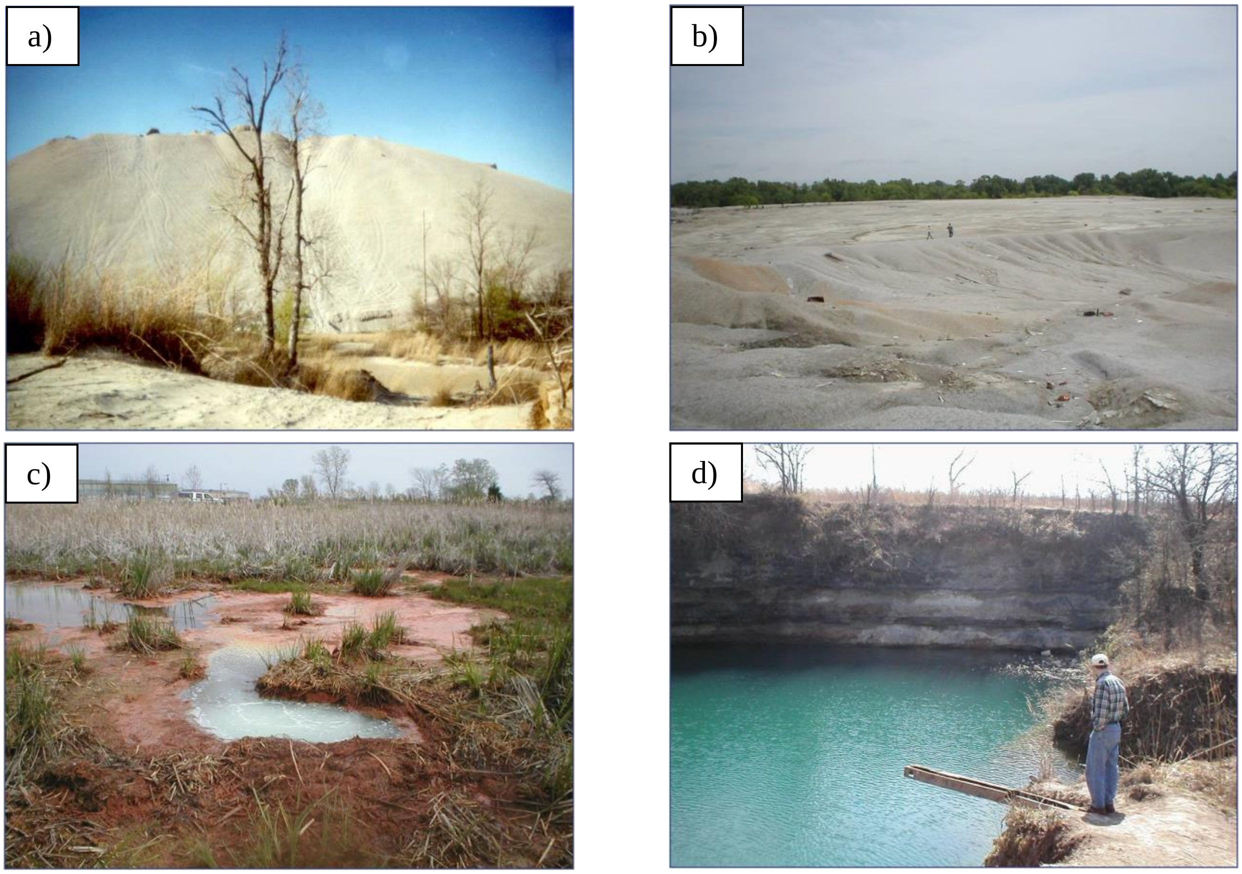

Figure 2. Landscape features of the Tar Creek watershed showing a) chat pile, b) fines pond, c) artesian discharge of mine water and d) surface collapse.

agency's ability to address other CERCLA releases (EPA, 1984, 1994, 2005, 2010). In the Third Five-Year Review of the OU 1 ROD (EPA, 2005), it was stated that " ... *EPA continues to find that, due to the potential drain on the Superfund and due to the impact that drain would have on the EPA's ability to address other releases under CERCLA... it is not appropriate to address environmental risks for surface water in Tar Creek.*"

Therefore, since November 1979, unmitigated flows of contaminated mine water, from both artesian mine pool discharges and chat pile/fines pond runoff and leaching, have impacted Tar Creek and its tributaries. The streams do not meet the pH standard and eco-toxic metals numeric criteria, even for the re-designated uses, over 30 years after first flush (e.g., Cope et al., 2008; LaBar et al., 2010).

Faculty, staff and students from the University of Oklahoma have focused environmental research and education efforts throughout the Tar Creek area and nearby watersheds since 1998 (e.g., Nairn et al., 2001; Coffey and Nairn, 2003; Iverson and Nairn, 2003; Nairn et al., 2005; White and Nairn, 2007; Winfrey et al., 2010; LaBar et al., 2010). Additionally, the University of Oklahoma Center for Restoration of Ecosystems and Watersheds (CREW) implemented a comprehensive watershed-scale environmental monitoring program from 2004-2010 (Nairn et al., 2008; Nairn et al. 2010b). In addition, CREW led an effort to design, construct and evaluate the first full-scale mine water treatment system in the mining district (Nairn et al., 2009, 2010a, 2010b). Evolution of watershed and mine pool water quality and recent advances in passive treatment technologies, coupled with land reclamation activities and socioeconomic changes, provide opportunities to revisit previous assumptions and reevaluate the potential to successfully address environmental risks posed by metal-contaminated mine waters.

Evolution of Watershed and Mine Pool Water Quality

Degraded water quality in streams due to mining influences has been documented in the Tar Creek watershed since at least the 1930s (Harbaugh, 1934; Haney, 1940; Ruhl et al., 1949). After cessation of major mining and pumping operations, significant artesian flows began to discharge in late 1979. These unabated artesian flows were the major drivers for use re-designation in 1985, although runoff and leachate have been shown to substantially contribute to degraded stream quality (e.g., Cope et al., 2008). The cumulative effects of these influences are reflected in in-stream water quality. Watershed-scale processes influencing stream water quality are complex both temporally and spatially, and include changes in ground-surface water interactions due to cessation of mining, chat-mining operations for asphalt aggregate (Wasiuddin et al., 2010), land reclamation activities and natural attenuation of contaminant loading.

Tar Creek near the US Highways 66/69 crossing (USGS Station 07185090 - Tar Creek near Commerce, OK) has been sampled periodically for decades (Table 1). Data from this location clearly illustrate the magnitude of these changes.

During active operations (~1914-1970), water associated with underground voids was continually pumped to Tar Creek and its tributaries (Harbaugh, 1934; Ledbetter, 1940). First flush occurred about a decade after all pumping ceased (November 1979) and prompted investigations by state and federal authorities (e.g., OWRB, 1981). With the determination that surface waters were irreversibly damaged, artesian discharge and runoff/leachate contributions continue to degrade water quality (CREW 2011).

Table 1. Selected water quality data for Tar Creek near US Highways 66/69 Bridge. Data are mean values (number of samples above detectable limits).

Source	Year(s) data collected	pH	Fe (mg/L)	Zn (mg/L)	Pb Mg/L)
Ledbetter (1940)	1940	6.47 (10)	1.06 (8)	111 (3)	0.13 (3)
OWRB (1981)	1980-1981	6.79 (10)	6.39 (12)	19.88 (12)	0.11 (12)
CREW (2011)	2004-2010	6.69 (72)	5.60 (62)	5.57 (62)	0.03 (34)

Given the carbonate geology of the mining district, pH remained circum-neutral throughout this period and statistical comparisons of the three sampling periods (1940, 1980-81 and 2004-2010) showed no significant difference for pH ($p>0.05$). In-stream iron (Fe) concentrations during the active mining period were relatively low as in-mine oxidation was limited by active water removal. Iron concentrations were greater in 1980-1981 and 2004-2010 when compared to the 1940 data ($p<0.05$ in both cases). The number of zinc (Zn) and lead (Pb) analyses from 1940 were limited making meaningful statistics comparisons impossible. However, Zn concentrations decreased from 1980-1981 to 2004-2010 ($p<0.05$). Lead concentrations did not differ between these sampling periods. Decades after cessation of mining and discharge of contaminated mine waters from the underground workings, in-stream metal concentrations remained elevated above applicable water quality standards (LaBar et al., 2010; Andrews and Masoner, 2011) yet Zn and Pb concentrations demonstrated a considerable decrease over time.

A major factor contributing to overall watershed water quality is the decrease in contaminant concentrations in artesian mine pool discharges since first flush. In large underground mine complexes, initial water quality associated with first flush is often the most contaminated and pollutant concentrations tend to decrease over time. Younger (2000) developed a simple relationship stating that the duration of first flush is approximately four times the duration of rebound (the time it takes for water to flood to the decant point), assuming the first flush is exponential. In this case, given that rebound duration in the Picher Field mine voids was 10-25 years (the majority of pumping operations ceased by 1955, with all pumping completed by 1970 and initial artesian discharges in late 1979), first flush would last approximately 40-100 years (through 2020-2080), after which discharge water quality would become asymptotic based on the rate of pollutant release near the zone of water table fluctuation.

Therefore, it is reasonable to expect decreases in contaminant concentrations in artesian mine pool water quality through time (e.g., when comparing 1980-1981 to 2004-2010 data). The initial points of discharge in 1979 were located at a site variously denoted as Commerce Springs, Site 14 (OWRB, 1981) and Seeps A and B (Nairn et al., 2010a; Nairn et al., 2010b). Comparisons of major metals concentrations for this site (Fig. 3a) demonstrate significant decreases in Al, Fe, Mn, Ni and Zn concentrations when 1980-81 data are compared to 2004-2010 data ($p<0.05$). However, comparisons of trace metal concentrations for these same two periods do not illustrate similar trends (Fig. 3b). Although Cu and Cr concentrations decreased, As, Cd and Pb concentrations increased.

Unfortunately, due to the almost complete lack of data between these two time periods (due to the re-designation of stream beneficial use and subsequent lack of monitoring), no long-term trend evaluations are possible. However, evaluation of the most recent data set does demonstrate significant decreases (Mann-Kendall Trend Analysis, $p<0.05$) for some metals over the six year sampling period. In addition to zinc, iron and manganese demonstrated significant decreases (Fig. 4). However, lead showed a significant increase over the sampling period.

Treatment of artesian discharges in the watershed was considered technically challenging and cost-prohibitive in the early 1980s (EPA, 1984) when only traditional active technologies (chemical dosing, mechanical aeration, etc.) were considered. Passive treatment technologies had not been widely explored nor implemented at that time (Huntsman et al., 1978; Weider and Lang, 1982) and were not considered an option in development of the OU1 Record of Decision. However, passive technologies advanced considerably in the late 20th and early 21st century and may represent a viable option of addressing surface water contamination in the Tar Creek and adjacent watersheds (Hedin et al., 1994; Watzlaf et al., 2004; Nairn et al., 2010b).

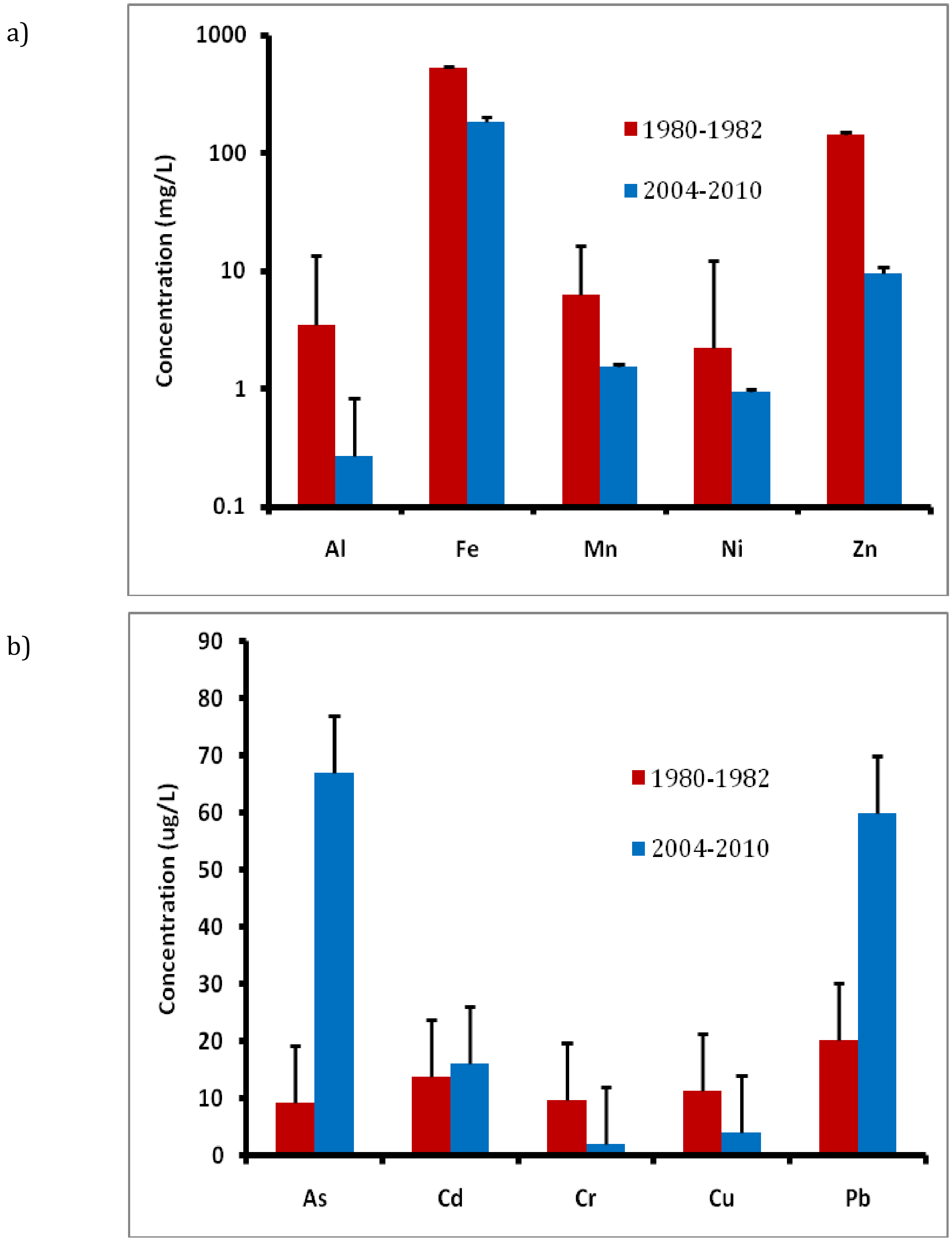

Figure 3. Comparison of a) major metal and b) trace metal concentrations for specific discharges for two distinct sampling periods shortly after first flush in 1979 and 25-30 years later near Tar Creek, OK.

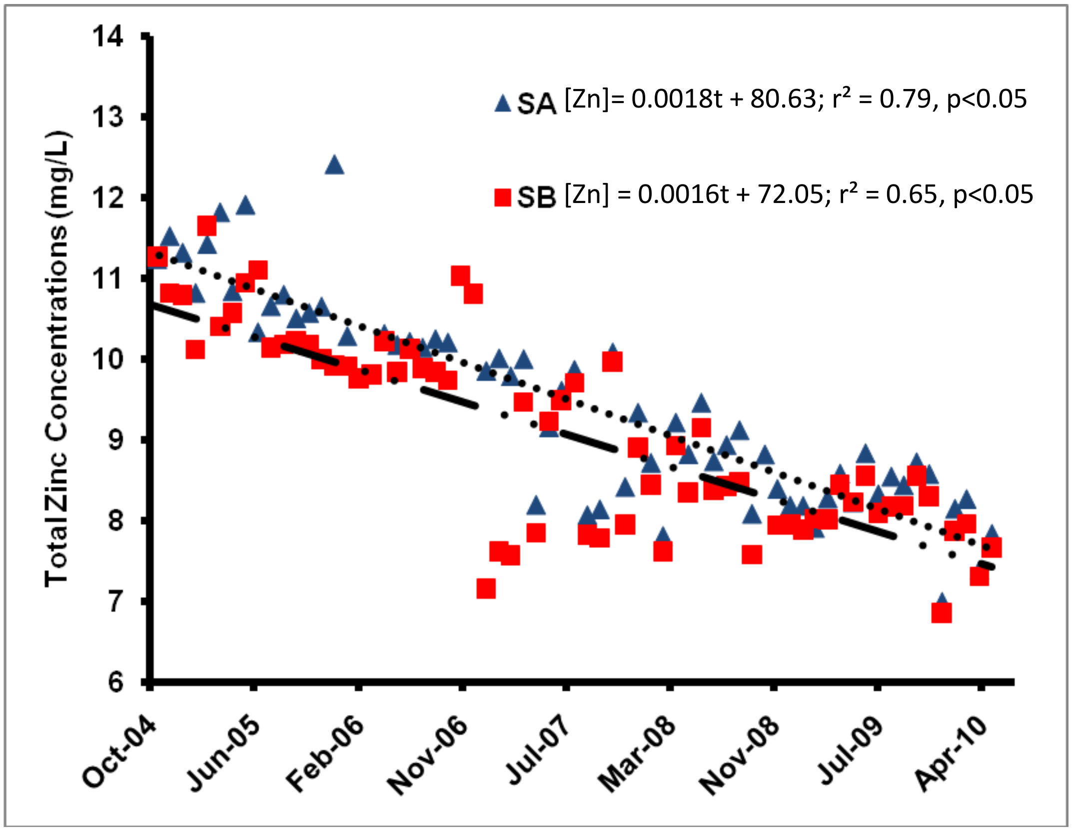

Figure 4. Zinc concentrations for selected discharges during the 2004-2010 sampling period near Tar Creek, OK, illustrating decreasing concentrations through time.

Passive Treatment Opportunities

In 2004, a project was initiated to design, construct and evaluate a full-scale passive treatment system for the Commerce Springs discharges described above. Design and construction was reported by Nairn et al. (2009) and initial evaluation documented by Nairn et al. (2010a, 2010b; Fig. 5 and Table 2). The system consists of ten distinct process units including two parallel treatment trains. Waters from three discharges (SA, SB and SD) flow into an initial oxidation pond (1), followed by parallel surface-flow aerobic wetlands/ponds (2N and 2S), vertical-flow bioreactors (3N and 3S), re-aeration ponds (4N and 4S), horizontal-flow limestone beds (5N and 5S), and are recombined in a single polishing pond/wetland (6). Each process unit is designed to carry out specific functions as described in Table 2 (Nairn et al., 2009).

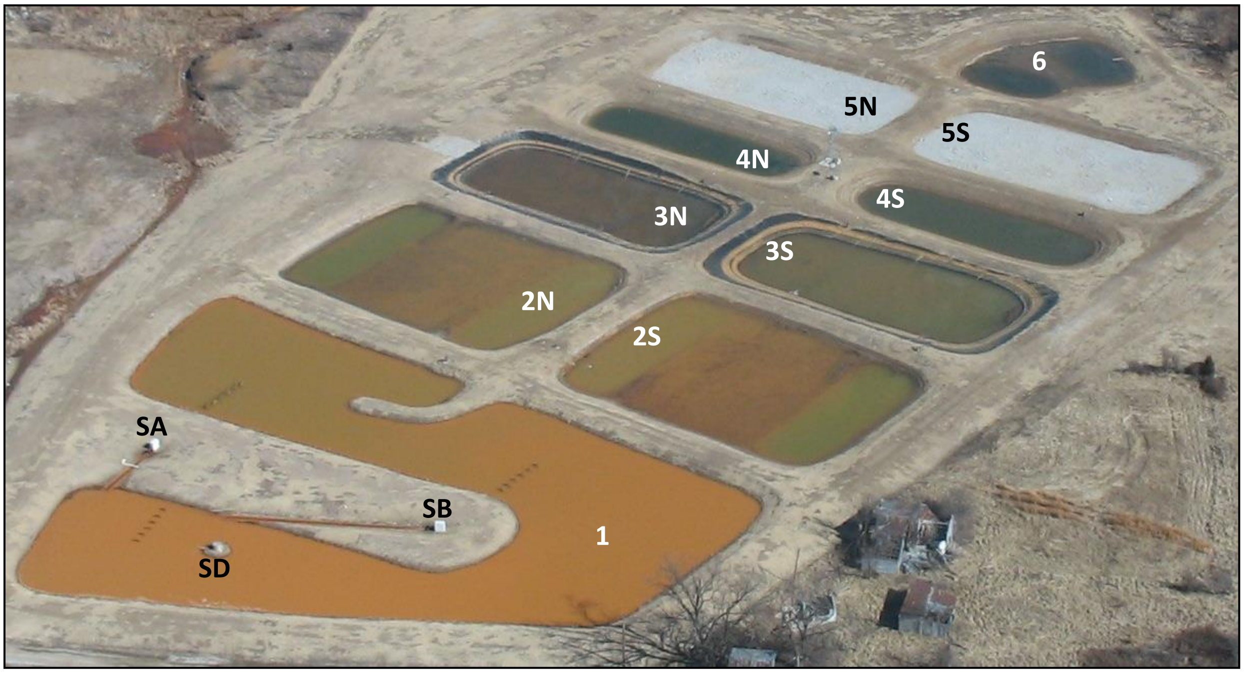

Figure 5. Oblique aerial photograph of passive treatment system taken showing mine drainage discharges and individual process unit designations near Tar Creek, OK.

In the first two years of operation, the passive treatment system met design specifications for water quality changes (Table 3). The net alkaline nature of the mine waters was maintained throughout the system as target metals were removed from solution. Due in part to degassing of elevated CO_2 concentrations in the artesian discharges, pH increased from < 6 to >7 at final discharge. Total Fe concentrations decreased to 0.57 ± 0.59 mg/L at the final outflow with the nearly 90% of total Fe removal occurring in the initial oxidation pond. Total Zn concentrations decreased to 0.10 ± 0.10 mg/L for the entire system, with removal occurring via sorption/co-precipitation in cell 1, sulfide precipitation in cells 3N and 3S, and $ZnCO_3$ formation in cells 5N and 5S. Total Cd, Pb, and As concentrations were decreased to below detection limits (0.64, 19.5 and 22 µg/L, respectively) before the outflow of the second process units, presumably through sorptive mechanisms. Approximately 95% of the Ni was removed via reductive mechanisms in the vertical-flow bioreactors, with final system effluent concentrations of 0.035 ± 0.02 mg/L.

Table 2. Summary of passive treatment system process units, primary targeted water quality parameters and design function, Tar Creek, OK.

Process unit	Targeted parameter	Function
Oxidation pond	Fe	Oxidation, hydrolysis and settling of iron oxyhydroxide solids; Trace metal sorption
Surface-flow wetlands/ponds	Fe	Solids settling
Vertical-flow bioreactors	Zn, Pb, and Cd	Retention of trace metal sulfides via reducing mechanisms
Re-aeration ponds	Oxygen demand and odor	Wind- and solar-powered re-aeration; Oxygen demand and H_2S stripping; Addition of O_2
Horizontal-flow limestone beds	Zn, Mn and hardness	Final polishing of Zn as $ZnCO_3$ and of Mn as MnO_2; Hardness addition to offset bioavailability of remaining trace metals
Polishing pond/wetland	Residual solids	Solids settling: Photosynthetic oxygenation; Ecological buffering

Table 3. Summary of total metals and sulfate concentration data for flow-weighted influent and final passive treatment system effluent, Tar Creek, OK. Data are presented in mg/L as mean ± standard error; BDL = below detection limits.

Parameter	Influent	Final effluent
Al	0.094 ± 0.009	0.071 ± 0.030
As	0.063 ± 0.002	BDL
Ca	742 ± 9.0	740 ± 22.3
Cd	0.016 ± 0.002	BDL
Co	0.066 ± 0.008	0.007 ± 0.0004
Cr	0.001 ± 0.0002	0.002 ± 0.0006
Cu	0.002 ± 0.0003	0.003 ± 0.0003
Fe	177 ± 2.33	0.57 ± 0.207
K	26.0 ± 0.286	31.1 ± 4.82
Li	0.366 ± 0.010	0.365 ± 0.018
Mg	200 ± 2.53	198 ± 7.49
Mn	1.51 ± 0.016	1.38 ± 0.197
Na	94.9 ± 1.63	96.6 ± 4.23
Ni	0.945 ± 0.015	0.035 ± 0.007
Pb	0.068 ± 0.003	BDL
Zn	8.29 ± 0.078	0.096 ± 0.037
SO_4	2239 ± 26	2047 ± 72

This passive treatment system represents the first full-scale mine water treatment system in the mining district. Additional artesian discharges are found in the upper Tar Creek and Beaver Creek watersheds (Fig. 1). However, contaminant concentrations and mass loadings are less than or similar to these untreated discharges (Table 4), making passive treatment an especially attractive option to address surface water quality degradation. At this time, the fund-balancing waiver regarding surface waters under the OU 1 ROD is still in place and any concerted effort to address water quality has not been undertaken. However, in the Fourth Five Year review of the OU 1 ROD, it was noted that passive treatment "*could be an economically feasible engineered remedy for surface water at the site. For these reasons, in this fourth five-year review, the fund balancing ARARs waiver included in the OU1 ROD may no longer be appropriate and should be reevaluated*" (EPA, 2010).

Table 4. Comparison of water quality characteristics for existing passive treatment system design and potential discharge to be treated in the Beaver Creek watershed, OK.

Parameter	Existing passive system design influent	Representative Beaver Creek artesian discharge
pH	5.95	6.62
Total alkalinity (mg/L)	393	185
Net alkalinity (mg/L)	29	90
Fe (mg/L)	192	9.28
Zn (mg/L)	11	1.60
Ni (mg/L)	0.97	0.02
Cd (mg/L)	17	1
Pb (mg/L)	60	15
As (mg/L)	64	20
SO_4^{2-} (mg/L)	2239	244
Discharge rate (LPM)	1000	25

Additional Factors

Additional factors that influence the feasibility of successful water quality remediation and stream restoration include ongoing land reclamation efforts (OCC, 2005; ODEQ, 2005), chat recovery for aggregate production (Wasiuddin et al., 2005, 2009), chat and fines injection into the mine pool (EPA, 2008), and a federally-funded buy out of the most at-risk mining communities (USACE, 2008).

By 1935, Netzeband (1937) reported that approximately 280 million metric tons of waste rock or tailings had been produced in the Tri-State Mining District and that a third had already been beneficially reused for aggregate, railroad ballast and other uses. By the early 21st century, approximately 68 million metric tons remained (e.g., OSE, 2000). State and federal guidelines for safe and effective reuse had been established (OSE, 2000; EPA, 2007) based on the work of Wasiuddin et al. (2005).

Although recovery for asphalt aggregate production is ongoing, residual fines and contaminated soils represent a considerable and continual environmental risk. Reclamation of the approximately 1000 ha of mining-disturbed land in the Oklahoma portion of the district has typically taken the form of waste removal and disposal activities (OCC, 2005). In 2008, a waste repository was established under Operable Unit 4 (EPA, 2008). This non-residential land reclamation effort (residential clean up was conducted under Operable Unit 2) is estimated to have a 30-year lifetime. In addition, unrecoverable fines materials are being actively injected into the underground void spaces as a disposal option (EPA, 2008). The long-term impact of injected fine material on mine pool water quality, and consequently artesian discharges, is unknown. However, as these source materials are removed and/or remediated, detrimental influences on stream water quality should be mitigated.

Although considerable funds were expended to remediate residential spoils under OU 2 in the 1990s and early 2000s, the mining communities in the center of the Picher Field have been the focus of two buy out and relocation efforts (ODEQ, 2006; USACE, 2008). In 2006, a state-funded effort was conducted for families with children six years of age and under, due to their vulnerability to neurological damage from lead exposure. After issuance of a subsidence report in 2006 (USACE, 2006), a federally-funded effort resulted in a nearly complete buy out and relocation of residents in the communities of Cardin, Hockerville and Picher. As of 2010, these communities have been dis-incorporated, their school districts closed, and nearly all commercial and residential structures have been removed.

In terms of human health risk, the relocation of residents essentially removes the receptor from proximity to the source of hazard. From an ecological risk perspective, the buy out and relocation efforts provide considerable opportunities for implementing large-scale land and water remediation and restoration projects in these watersheds, if adequate funding is provided over an appropriately planned time period.

From a water quality improvement perspective, given an as yet unrealized understanding of mine pool hydrology, seven related watershed remediation/restoration options should be considered: i) permanent closure of appropriate locations of surface/mine pool interaction, ii) passive treatment of remaining artesian mine pool discharges, iii) passive treatment of remaining chat pile/fines pond leachate/runoff after land reclamation activities have been completed, iv) restoration of stream channels through chat sediment removal and re-establishment of fluvial geomorphologic connectivity, v) creation and restoration of wetland habitat both in riparian and other appropriate areas and vi) establishment of native prairie habitat in uplands and vii) institution of an appropriately holistic long-term monitoring and data archiving effort for adaptive management.

Conclusions

A century of hard rock mining in the Tri-State Mining District has resulted in substantial environmental degradation of land and water resources. Although the region was designated as a suite of EPA Superfund sites in the early 1980s, streams draining the mining-impacted watersheds still fail to meet designated beneficial uses, in part due to administrative decisions rendering them "irreversibly damaged". However, three decades of remediation work, coupled with changes in mine pool water quality and advances in passive treatment technologies, merit reconsideration of these decisions. Furthermore, given the recent relocation of residents associated with the most severely impacted communities, unique opportunities exist for large-scale ecological engineering implementation projects that could effectively address water quality. Initial results from a pilot passive treatment system are promising and fit well with possible future land uses being considered by the Quapaw Tribe of Oklahoma (i.e., a return to native prairie and associated wetland and riparian habitats). Successfully addressing water quality higher in the watersheds would also provide substantial downstream benefits to other Native American tribes, residents and fish and wildlife resources.

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