Estimation of Groundwater Residence Time in Tailings from the Elizabeth Copper Mine Superfund Site, Vermont, USA

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Abstract

Concentrations of several environmental tracers were measured in water within the tailing piles at the abandoned Elizabeth copper mine to estimate the residence time of groundwater and aid in the evaluation of long-term water treatment options. Current remediation plans include surface-water and shallow groundwater diversion, installation of drains, regrading, capping, and passive treatment of residual discharge. Apparent groundwater ages were estimated from tritium, sulfur-hexafluoride, chlorofluorocarbon (CFC), and tritium-helium data, and water-balance calculations. The pre-existing discharge chemistry from the tailings was characterized by near-neutral, net acidic, ferrous sulfate waters, containing several g/L TDS. Tritium concentrations indicate an apparent age no older than ~1970 for recharge of the groundwaters. The range of apparent CFC ages suggests degradation of CFCs, consistent with the anoxic environment of the sulfidic tailings. The remaining dating techniques indicate a groundwater velocity between 16 and 33 m/yr, which is consistent with estimates from laboratory and field measurements of hydraulic conductivity. However, water-balance calculations based on discharge from the base of the pile indicate a significantly faster rate of replenishment of the pore volume of approximately two years. This observation suggests that some groundwater may, in part, be utilizing fast channelizing flow paths through part of the tailings while the main mass of water is traveling through the saturated pore space.

Key Words: dating, CFC, sulfur hexafluoride, tritium-helium, pyrrhotite, mass balance

Introduction

The Elizabeth copper mine in east-central Vermont operated intermittently from 1809 until 1958. The mine worked a north-plunging, copper-rich pyrrhotitic massive sulfide ore body by both open pit and underground mining methods (Hammarstrom *et al.* 2001; Kierstead 2001). Early mining focused on the production of copperas ($FeSO_4 \bullet 7H_2O$, also known as melanterite) through the roasting and heap-leaching of pyrrhotitic massive sulfide, followed by evaporation of the leachate in vats to crystallize the copperas. Subsequent early production focused on copper recovered from hand-sorted ores, which were smelted on site. Mine wastes from these early operations were found in a series of waste piles known as TP3 in the headwaters of Copperas Brook, a small tributary of the West Branch of the Ompompanoosuc River (Figures 1 and 2). From 1942 to 1958, mining used modern underground mining and ore-processing methods to produce a chalcopyrite concentrate using froth flotation. The flotation tailings were deposited in two piles, known as TP1 and TP2, totaling roughly 3 million tonnes (Mt), downstream of TP3 in the Copperas Brook valley.

The U.S. Environmental Protection Agency placed the Elizabeth mine on its National Priorities ("Superfund") List in 2001 due to acid-mine drainage and its downstream ecological impairment of Copperas Brook and the West Branch of the Ompompanoosuc River. Impairment is observed in both water quality and impacts to benthic macroinvertebrate and fish communities for several kilometers downstream. Groundwater in the vicinity of the waste piles exceeds drinking water standards for Cd, Cu,

Mn, and Ni (URS Corporation 2009). Additional concerns included the physical instability of tailings pile TP1 and the associated risks to residents living downstream along the West Branch. The adjacent Lord Brook watershed to the southeast of the main mine workings contains a small pit lake, shallow underground workings, and several waste-rock piles that will be included in the remedial action.

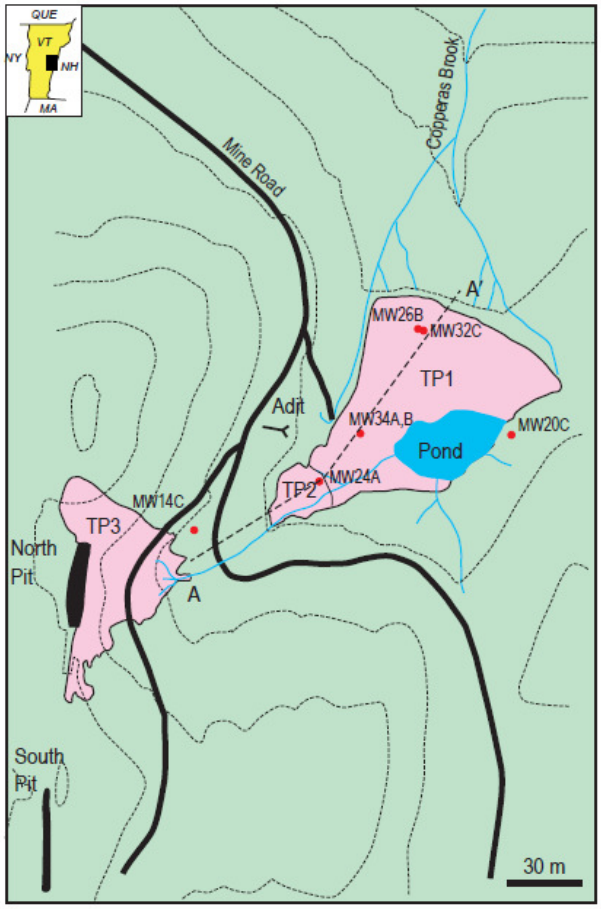

Figure 1. Schematic map showing main features of the abandoned Elizabeth mine site prior to remediation. The locations of monitoring wells investigated in the study are shown as red dots. The location of the cross section shown in Figure 3 is indicated by the dashed line labeled A – A'.

Remediation within the Copperas Brook watershed began in 2003 and is still ongoing. It includes:

- Construction of diversion trenches upgradient of TP1 and TP2 to reroute Copperas Brook and intercept shallow groundwater
- Construction and operation of a temporary active treatment system to treat acidic, ferrous sulfate groundwater discharging from the base of TP1
- Relocation of TP3 atop TP1 to eliminate the source of acid-mine drainage in the headwaters of Copperas Brook
- Plugging of historical decant system within TP1
- Regrading of the combined TP1, TP2 and TP3 to stabilize the tailings piles and facilitate surface-water drainage
- Installation of toe drains and horizontal drains to lower water table within TP1
- Placement of a low-density polyethylene cover overlain by a 0.6 m revegetated (grass) soil cover to eliminate the infiltration of precipitation into the combined tailings pile

- Decommissioning of the active treatment system and replacement by a yet-to-be-determined passive treatment system.

Figure 2. Aerial photograph of the tailing piles at the abandoned Elizabeth looking south circa 2007. TP1 is in the foreground. The brown slope break at the back corner of TP1 is TP2. The bottom end of TP3 is barely visible as the brown area at the right edge of the photograph. Copperas Brook emerges from TP3, is currently diverted along the eastern (far) edge of TP1, then follows through a channel along the northern (near) face of TP1 until it reaches its natural channel.

The final design of the passive treatment system presents a number of challenges because of the limited space afforded by the short (< 1 km), steep and narrow stream valley below TP1 before the confluence with the West Branch. An accurate knowledge of the residence time of groundwater in TP1 is required to enable effective design of the passive treatment system and ensure successful, long-term treatment. This study employed a variety of approaches to assess the residence time of groundwater within TP1. These approaches include tritium dating, chlorofluorocarbon (CFC) dating, sulfur-hexafluoride (SF_6) dating, tritium-helium dating, and water-balance calculations, all of which can be compared to various field and laboratory hydrologic measurements (URS Corporation 2009).

Study Area Description

The two tailing piles at the Elizabeth mine were constructed using upstream methods on a substrate of glacial till and alluvium. Dams were built of tailing along the northern and western sides of TP1 – the larger of the two piles – and along the northern end of TP2. Accordingly, the grain size of the tailings decreases away from the dam structures. The median grain size ranges between silt and fine sand. The crest of TP1 was approximately 37 m above its base; the crest of TP2, which was constructed at the southern end of TP1, was approximately 15 m above its base (Figure 3). The total combined volume of the two tailings piles is 1,835,000 m^3 (URS Corporation 2009).

Both TP1 and TP2 were drained by separate decant systems that included a vertical pipe with the elevation maintained near the top of the pond during construction joined to a horizontal discharge pipe at the northern end of the pile. The TP2 decant system failed in the 1970s. The TP1 decant system was plugged during the current remediation. In addition, TP1 was also drained by a series of horizontal drains

emerging from the base of the pile, along its northern edge. Historically, these discharges, like the groundwater within the pile, were characterized neutral-pH, net acidic, ferrous sulfate waters, containing several grams per liter total dissolved solids (Table 1; Seal *et al.* 2001). During regrading, a toe-drain system was installed along the northern face of TP1. Throughout most of the year, effluent from the toe-drain and horizontal drain system is collected and pumped to an active treatment system before being discharged into Copperas Brook.

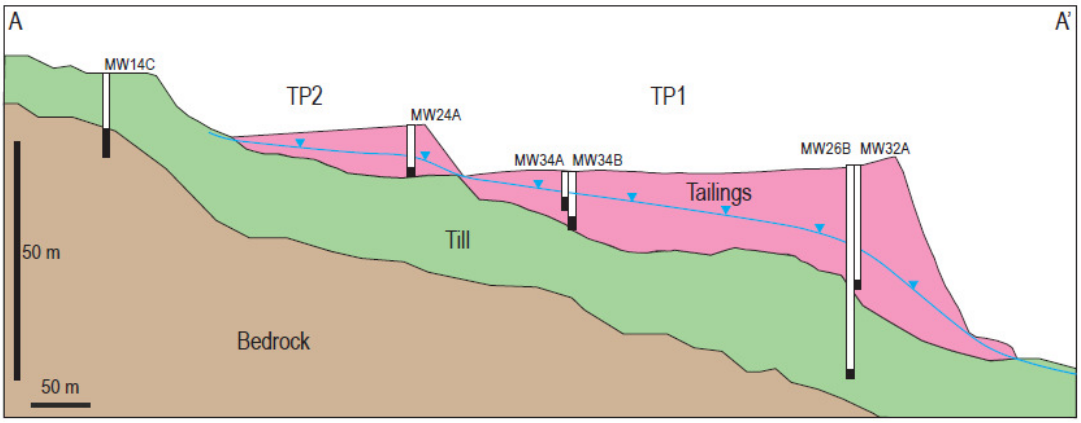

Figure 3. Cross section of the Elizabeth mine tailing piles. The location of monitoring wells sampled (white bars) in the study and their screened intervals (black bars) are shown. See Figure 1 for the location of the cross section. Note: 10x vertical exaggeration.

The Elizabeth mine experiences a temperate, humid climate. The average annual precipitation is 98.3 cm and is evenly distributed throughout the year. The average temperature in January, the coldest month, is -7.5 °C, and the average temperature in July, the hottest month, is 21.5 °C.

The depth to the water table (phreatic surface) historically remained fairly constant throughout the year. Within TP2, the phreatic surface was near the base of the pile due to the failure of the decant system. Within TP1, the phreatic surface was within 0.5 m of the surface of the pile at the southern end near the base of TP2, but was found at a depth of approximately 19 m near the crest of the pile. Approximately 24 percent of the pile is saturated (URS Corporation, 2009).

Table 1. Geochemical characteristics of groundwater samples.

Sample	Water T °C	pH	S.C. mS/cm	Eh mV	Alkalinity mg/L $CaCO_3$	Fe mg/L	SO_4 mg/L
MW14C	11.0	6.9	0.581	356.7	76.8	<0.02	233.1
MW20C	10.8	8.0	0.138	346.5	50.8	<0.02	13.1
MW24A	9.3	6.6	1.034	159.9	140.4	77.50	546.2
MW26B	12.0	7.4	1.291	90.9	134.4	0.11	635.2
MW32C	15.9	7.0	3.450	-130.0	154.0	25.40	2,383.3
MW34A	9.6	5.8	2.970	238.0	84.8	447.0	3,118.0
MW34B	10.1	5.3	3.060	330.5	8.2	506.0	3,617.3

Samples, Methods, and Results

A total of 35 water samples were collected for tritium analysis from monitoring wells and surface-water sources around the site. A subset of seven samples from monitoring wells was collected in October 2009 for CFC, SF_6, tritium-helium (^3H-^3He), dissolved gas, and noble gas analysis. These samples included two bedrock wells upgradient of TP1 and TP2 (samples MW-14C and MW-20C), one sample of

groundwater from the till beneath TP1 (MW-26B), and four samples of groundwater within TP1 (MW-32C, MW-34A, MW-34B) and TP2 (MW-24A) (Figures 1 and 3). The CFC and SF_6 samples were collected in bottles immersed in a bucket of sample water at atmospheric pressure. The tritium-helium samples were collected in crimped copper tubes mounted in a manifold that could be overpressured to keep dissolved gases in solution. The one sample for which overpressuring was not possible during collection due to gritty tailings fouling the valves was MW-32C from the northern crest of TP1. Dissolved gas and noble gas analyses are summarized in Table 2. Tracer analyses (CFC-11, CFC-12, CFC-113, SF_6, radiogenic helium ($^3He_{tri}$), and tritium are summarized in Table 3. Apparent ages based on the CFC, SF_6, and ^3H-^3He dating are summarized in Table 4.

Table 2. Gas chemistry of wells sampled for dating.

Sample	CH_4	CO_2	N_2	O_2	Ar	He	H_2	Ne
	mmol/L	mmol/L	mmol/L	mmol/L	mmol/L	nmol/L	nmol/L	nmol/L
MW14C	0.00010	0.22783	0.77957	0.00799	0.01820	4.14	7.42	16.50
MW20C	0.00000	0.06498	0.75878	0.10590	0.01892			
MW24A	0.00768	1.38723	0.66219	0.00707	0.01650	3.44	2.69	13.3
MW26B	0.00039	0.17446	0.68671	0.00787	0.01698	3.99	4.50	11.82
MW32C	0.00707	0.65472	0.61818	0.00765	0.01685	3.35	2.22	11.90
MW34A	0.02263	4.55085	0.59784	0.00641	0.01630	3.60	2.42	12.06
MW34B	0.00670	4.27550	0.65546	0.00701	0.01798	4.06	1.75	12.68

Table 3. Tracer concentrations of wells sampled for dating.

Sample	CFC-11	CFC-12	CFC-113	SF_6	$^3He_{tri}$	^3H	Noble Gas Recharge	Excess Air
	pmol/kg	pmol/kg	pmol/kg	fmol/kg	T.U.	T.U.	T °C	ccSTP/L
MW14C	0.451	0.743	0.072	1.733	34.00	5.0	12.6	5.9
MW20C	2.648	2.765	0.454	2.467	14.75	7.0	6.9	2.9
MW24A	0.116	0.444	0.011	2.732	5.02	5.0	9.7	3.1
MW26B	0.348	105.7	0.161	1.083	33.21	5.0	12.9	3.1
MW32C	0.230	1.698	0.070	0.674	3.85	5.0	8.3	-0.3
MW34A	0.152	8522.5	0.010	2.115	1.32	5.0	9.5	-0.4
MW34B	0.123	358.8	0.014	2.125	2.53	5.0	5.5	-0.6

Table 4.Apparent average recharge years (before 2009)*.

Sample	CFC-11 CE	CFC-11 UA	CFC-113 CE	CFC-113 UA	CFC-12 CE	CFC-12 UA	SF_6 UA	SF_6 CE	^3He-^3H
MW14C	45.1	45.1	34.3	33.9	38.8	38.4	12.8	13.5	40.2
MW20C	35.5	35.3	22.5	22.3	22.8	22.8	9.5	8.0	27.6
MW24A	53.3	53.3	48.8	49.0	43.3	43.8	5.0	-0.2	15.0
MW26B	46.9	46.8	27.9	27.8	Cont.	Cont.	19.3	16.8	53.6
MW32C	50.0	50.0	36.0	36.0	31.0	31.5	25.8	23.0	16.3
MW34A	52.3	52.0	50.8	50.8	Cont.	Cont.	10.8	4.3	5.3
MW34B	54.3	54.0	48.8	48.8	Cont.	Cont.	13.3	9.3	9.2

*CE and UA refer to models for accounting for the effect of excess air on calculated ages; “Cont.” is contaminated, meaning that the concentration exceeds that of air-saturated water.

Both CFC and SF_6 dating techniques are based on monotonic increases in atmospheric concentrations of these compounds as industrial atmospheric contaminants and the ability to relate these concentrations to recharge ages (Plummer and Busenberg 1999; Busenberg and Plummer 2000). However, international

regulation of CFC use has caused atmospheric concentrations to decrease in recent years, causing ambiguity in the interpretation of results from young waters. CFC dating utilizes three different CFC compounds: CFC-11, CFC-12, and CFC-113. Tritium-helium dating, unlike traditional tritium dating, is based on a parent-daughter isotopic ratio. Tritium (^3H) decays to ^3He with a half-life of 12.32 years; this decay forms the basis for this dating technique (Solomon *et al.* 1993). Tritium is an inherent part of the water molecule, but helium is a dissolved gaseous species.

Tritium

Tritium analyses can be used to assess the age of waters. Tritium is produced by both upper atmosphere cosmic ray interactions with atmospheric nitrogen, at low levels, and by thermonuclear explosions. "Bomb" tritium in the atmosphere has varied with the amount of nuclear testing as shown by long-term monitoring at various sites around the world. Nuclear testing reached a peak in the mid-1960s and mostly stopped by 1980. Thus, most atmospheric tritium has now decayed to near "cosmogenic" concentrations. Tritium values for waters at Elizabeth show limited variation, all centering around "modern" values with analytical uncertainty for the tritium values ranging from 0.3 to 1.3 tritium units (T.U.) with an average uncertainty of 0.6 T.U. (Figure 4), suggesting "recent" recharge for these waters (i.e., post-1960s). The only exceptions are the samples with low values (< 5 T.U.), P-3 located at the base of TP1 below the starter dam, MW-13B and C located at the base of TP3 on the north side of Mine Road, and MW-14C located at the base of TP3 west of the bend in Mine Road at Copperas Brook. These three samples likely represent mixtures of recent waters with "old" water (i.e., pre-bomb peak, recharged prior to 1960).

CFCs

The interpretation of CFC (and SF_6) data requires a number of parameters related to the partitioning of gases at the atmosphere-water interface. These parameters include estimates of recharge temperature and elevation, and the amount of "excess air" incorporated into the water during entrainment in groundwater flow systems. The amount of "excess air" is evaluated using two different models to account for the effect of "excess air" and concentrations of dissolved gases (Table 3). This "excess air" is generally interpreted to represent gas bubbles trapped in the vadose zone near the water table that become dissolved in the groundwater. The two most prominent are the CE and the UA models (Plummer and Busenberg, 1999; Aeschbach-Hertig *et al.* 2000); neither offers clear advantages over the other, and both yield similar recharge ages for the Elizabeth groundwaters (Table 4). The low excess air values for the TP1 and TP2 samples are consistent with the limited fluctuations historically observed in the water table within the tailings piles. CFC samples are also prone to contamination from anthropogenic sources, which can invalidate their use as a dating tool.

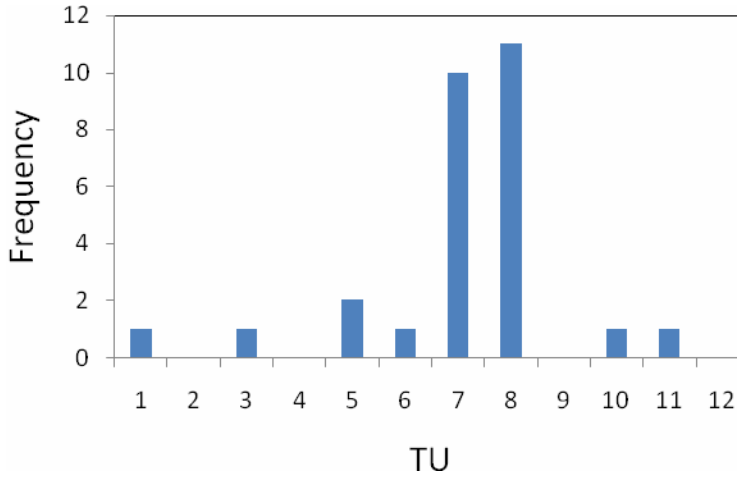

Figure 4. Histogram of tritium analyses for groundwater in the vicinity of the tailing piles at the Elizabeth mine.

In general, the CFC recharge ages are consistent with one another. Invariably, the CFC-11 apparent recharge ages are older than those based on either CFC-12 or CFC-113 (Figure 5). This observation is consistent with the anaerobic, methanogenic conditions within TP1 and TP2. CFC-11 is known to be the most susceptible to microbial degradation and CFC-12 is the least susceptible (Plummer and Busenberg, 1999). Due to anaerobic, methanogenic conditions within TP1 and TP2, the CFC ages that are most likely to be meaningful are those from the bedrock wells upgradient from TP1 and TP2 (i.e., MW-14C and MW-20C). Here, good agreement is found among the CFC-12, CFC-113, and tritium-helium ages (Table 4). In other words, the groundwater at MW-14C was recharged around 1970, and that at MW-20C was recharged between 1982 and 1987. Note, however, that the CFC results (Table 1) indicate that sample MW-14C may represent a mixture of "recent" water and "old" water.

SF_6

The interpretation of SF_6 data requires similar considerations as CFCs in terms of excess air, and recharge temperature and elevation. The industrial use of SF_6 began in 1953; thus, its usefulness as a groundwater dating tool is generally limited to waters recharged since 1970 (Busenberg and Plummer, 2000). Unlike CFCs, it does not degrade or adsorb on to solids. Sulfur hexafluoride is more prone to geologic contamination than CFCs, but the risk of geologic contamination at the Elizabeth site is minimal because of the absence of problematic rock types. Invariably, the apparent SF_6 ages are younger than the corresponding CFC or tritium-helium ages with the discrepancy being greatest at the older apparent ages (Figure 5). Part of the explanation for the discrepancy in the older ages may be that the inferred ages for these samples are near the operable limits for the SF_6 technique, and probably are not reliable.

Tritium-helium

Tritium-helium dating, unlike traditional tritium dating, is based on a parent-daughter isotopic ratio. Tritium is an inherent part of the water molecule, but helium is a dissolved gaseous species. Thus, partial degassing of samples is a potential source of error in age determinations that produces an erroneous young bias. Problems related to degassing can be overcome by overpressuring the sample container during collection to keep dissolved gases in solution. The one sample for which overpressuring was not possible during collection due to gritty tailings fouling the valves was MW-32C from the northern crest of TP1. Tritium-helium ages for the bedrock wells upgradient from TP1 and TP2 agree well with the CFC ages as discussed above (Table 4). The oldest tritium-helium age (MW-26B, 53.6 years) is from the till beneath TP1 (Figure 3). From a hydrologic perspective, this could be the oldest water sampled. In contrast, the younger apparent tritium-helium ages agree reasonably well with the younger SF_6 ages, but are distinctly younger than the CFC ages (Figure 6).

Water balance

An alternative approach for evaluating the residence time of water in the tailing piles is to consider the mass balance of water entering and exiting the pile. Discharge from the toe drain and horizontal drains at the base of TP1 has been carefully monitored since the construction of the temporary active treatment system in 2007. The till at the base of the pile effectively acts as an aquitard. Therefore, discharge to the local aquifer should be minimal. Thus, based on the calculated volume of pore water and the measured discharge, the entire saturated volume of the tailing pile should be drained in approximately two years (Table 5). The annual water flux from the drains is roughly equivalent to the annual amount of precipitation on the combined tailings pile. However, only a fraction of the precipitation actually infiltrates the pile.

Discussion

The recharge age estimates based on gaseous species are susceptible to a number of complicating factors. Gaseous species can outgas under conditions of warmer temperatures or lower pressures than those found in the aquifer. In the case of CFCs and SF_6, the partial stripping of dissolved gases would bias the interpretation towards erroneously old ages. In the case of tritium-helium, the daughter product would be stripped, which would lead to a bias towards erroneously young ages. In the case of the CFC and SF_6 samples, water was pumped into sample bottles immersed in a bucket at atmospheric pressure. Outgassing due to warming is unlikely, but outgassing due to decompression is more likely, especially because the samples contained considerable concentrations of dissolved gases (Table 2). For tritium-helium analysis, the samples were collected in a manifold that could be overpressured, which would minimize outgassing.

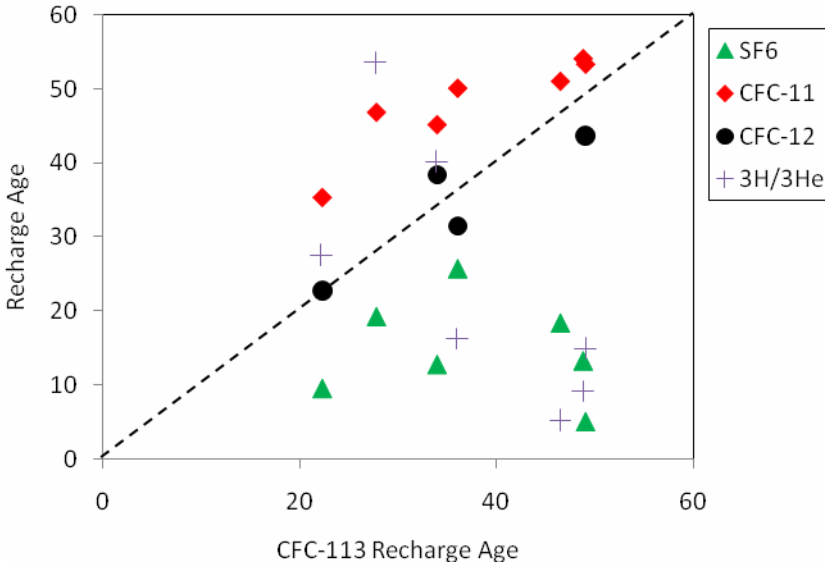

Figure 5. Apparent ages for groundwater samples based on various techniques relative to the CFC-113 apparent ages (UA model). The dashed line represents ideal agreement among the ages.

Table 5. Water-balance calculations for tailings water budget.

	Parameter
Combined tailings volume	1,834,932 m^3
Surface area	113,310 m^2
Annual precipitation on tailings surface	111,045 m^3
	111,045,000 L
Porosity (assumed)	50 %
Pore volume	917,465,830 L
Saturated percent	24
Saturated pore volume	220,191,799 L
TP1 average drain discharge	201.4 ± 20.0 L/min
TP1 annual mean drain discharge	105,847,381 L
Replacement Period	2.1 years

An additional complicating factor with CFC and SF_6 dating is that the gas partitioning is assumed to occur under atmospheric conditions at the phreatic surface. While this assumption may be valid for the bedrock samples, it may not be valid for recharge within the tailing piles. Pyrrhotitic tailings such as those at Elizabeth rapidly consume oxygen due to sulfide oxidation (Blowes *et al.* 2003). Thus, the atmosphere at the phreatic surface within the tailing piles is very likely devoid of oxygen. Corrections for

excess air and other trace gas may not be straightforward because some gaseous species, such as CO_2 and CH_4, may be evolved in the flow path rather than being entrained at the phreatic surface. The dissolved CO_2 is likely produced from the dissolution of trace calcite within the tailings pile (Blowes *et al.* 2003, Seal *et al.* 2001).

The validity of various estimates of groundwater velocity can be evaluated by comparing estimates made by various means. The distance between MW-34B and MW-32C is approximately 230 m. This distance, combined with recharge age estimates, allows groundwater velocity to be estimated (Table 6). These estimates can be compared to estimates based on simple Darcy's Law calculations using lab and field hydraulic conductivities determined by URS Corporation (2009). Lab hydraulic conductivities for tailings ranged from a low of 1.4×10^{-7} cm/s to a high of 1.0×10^{-3} cm/s. The large range of lab hydraulic conductivities reflects the grain size differences between the coarse tails near the discharge points and the fine grained slimes closer to decant points. Interlayering of coarse and fine tails is expected to produce anisotropy in pile with lower vertical and higher horizontal conductivities. The average hydraulic conductivity from the slug tests performed in the field was 7.1×10^{-4} cm/s, which is similar to the higher values observed in the lab.

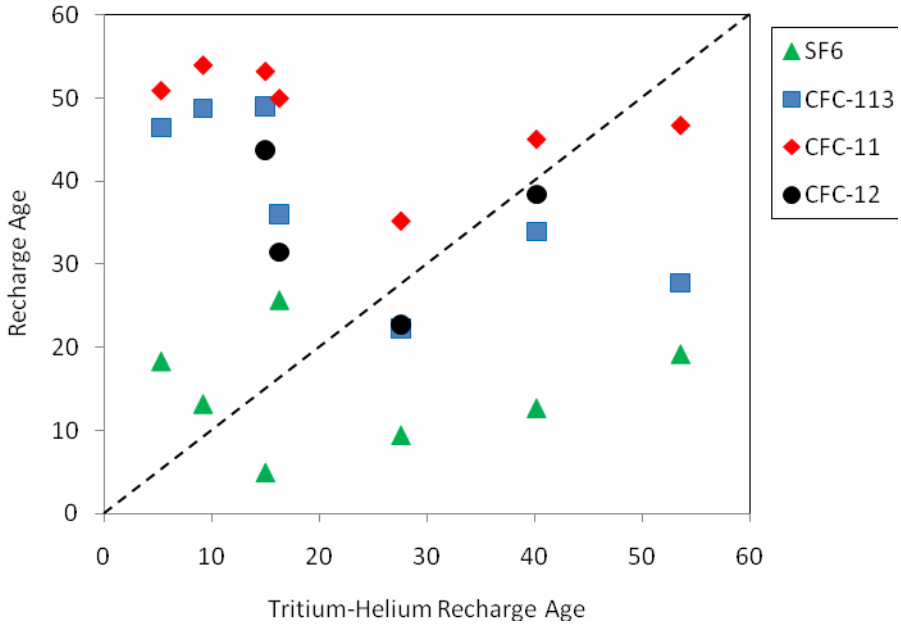

Figure 6. Apparent ages for groundwater samples based on various techniques relative to the tritium-helium apparent ages. The dashed line represents perfect agreement among the ages.

Table 6. Comparison of estimates of groundwater velocity.

Method	Velocity (m/yr)
SF_6 UA	18.3
$^3H/^3He$	32.3
Lab hydraulic conductivity (maximum)	36.2
Lab hydraulic conductivity (minimum)	0.01
Field hydraulic conductivity (slug ave.)	25.6

This comparison indicates that the velocity estimated on the basis of the SF_6 ages is approximately 30 percent slower than the estimate based on the average slug-test hydraulic conductivity. The velocity based on the $^3H/^3He$ ages is significantly faster, but undoubtedly incorrect because of the sampling

problem for sample MW-32C. In any case, the velocity estimated based on the SF_6 and tritium-helium dating is within the range of those estimated on the basis of hydraulic conductivities measured in the laboratory on Elizabeth mine tailings, and that estimated from field slug tests.

A remaining perplexity of the Elizabeth mine tailings hydrology is the fact that a comparison of the discharge from the toe and horizontal drains with the volume of pore water within the pile indicates that the replacement period for the entire volume of pore water within the tailings pile should be on the order of 2 years (Table 5). This rate is significantly faster than the velocities estimated on the basis of the various dating techniques and those estimated with calculations using measured hydraulic conductivities. This conclusion suggests that the flow within the pile consists of two components: slower base flow through the pore volume of the saturated zone, and faster, channelized flow, possible utilizing the premining stream channels of Copperas Brook and its tributaries that have been covered by tailings. The premining channels may represent the locations of sources of groundwater influx to the piles, because these channels are the most likely areas where the impermeable glacial till may have been breached.

In summary, abandoned mine tailing piles present myriad challenges for estimating the residence time of associated groundwaters. The anoxic environment found in the vadose zone of sulfidic tailings probably requires modification of conventional modeling of atmosphere-water partitioning of trace gases. Likewise, chemical reactions between acid-mine drainage and carbonate minerals within the tailings present challenges for sampling and may further complicate interpretation of analytical results under the best of conditions. Reasonable estimates of the residence time of water are best obtained by comparing multiple lines of evidence and constrained by a well defined water balance. Future work at the Elizabeth mine will seek to understand better the magnitude of these complicating factors in the dating of groundwaters in sulfidic tailing piles.

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Acronyms

CFC	chlorofluorocarbon
TDS	total dissolved solids
Mt	million tonnes
S.C.	specific conductance
mmmol	millimole
nmol	nanomole
pmol	picomole
fmol	fentomole