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Attenuation of acid rock drainage from buffering by naturally occurring calcite at the Leviathan Sulfur Mine, California

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Abstract

The former Leviathan Sulfur Mine is located within the East Fork Carson River drainage of California and Nevada. The sulfur was deposited as part of a high sulfidation epithermal system common to the Walker Lane structural belt. Acid rock drainage (ARD) occurs at the site and has impacted groundwater and surface water. The oxidation of pyrite and, to a lesser extent arsenopyrite and sulfur, within hydrothermally altered ore rock and mine spoil (waste), is the source of ARD. The ARD is characterized by low pH, and high sulfate and metals concentrations.

Calcite is present within unaltered country rock, comprised of volcanic and volcaniclastic sedimentary units, outside of the hydrothermally altered core and ore deposit area. The calcite has been mapped as occurring as fracture and vug-filling cement within lahars and sandstones, and filling fractures and vugs in basalt and andesite. In addition, a downslope sandstone outcrop containing fracture-filling calcite has been observed and documented. The presence of calcite was further confirmed by x-ray diffraction analysis of rock samples from boreholes within sedimentary units downslope (downgradient) of the mine; and was additionally confirmed in unaltered rock in 25 boreholes surrounding the altered area.

A literature review showed calcite is commonly found in such epithermal deposits throughout the region. The calcite precipitates as platy crystals filling voids or crusts filling fractures on the periphery of shallow alteration zones at temperatures generally less than 180^0 C. The precipitation occurs through exsolution of CO_2 in boiling fluids from the volcanic-hydrothermal system.

Sampling and analysis of groundwater from monitoring wells shows evidence of buffering of ARD by calcite vertically and laterally downgradient of the mine. This included an increase in bicarbonate and pH coupled with decreasing metals and metalloid concentrations in wells that had calcite confirmed in the boreholes, compared to well boreholes without calcite. The mapped extent of ARD also shows the vertical and lateral extent is limited by the pervasive occurrence and buffering by calcite in bedrock underlying the shallower unconsolidated unit.

Keywords: *acid drainage, calcite, pyrite, natural attenuation*

1 Introduction

The Leviathan Mine is a former underground and open-pit sulfur mine located in a remote mountainous area of northeastern Alpine County, California, about 25 miles southeast of South Lake Tahoe (Figure 1). The mine is positioned on the eastern slope of the central Sierra Nevada at approximately 2,130 meters above mean sea level (amsl) and lies within the East Fork Carson River drainage of California and Nevada. Open pit operations for sulfur mining were conducted at the site from late 1952 until abandonment in 1962. During this time, over 20 million tons of overburden and waste rock containing sulfide minerals were removed from the pit area and distributed throughout the mine site.

The mine encompasses approximately 102 hectares of land disturbed by historical mining activity and subsequent environmental response actions. The key physical features of the mine are shown on Figure 2 and include: an open pit present near the centre of the mine; a collapsed portal to an adit (Tunnel 5) near the pit; predominantly regraded spoil (mine waste) piles located roughly to the north, south and west of the pit; a surface discharge collection system including five settling ponds; a pit clarifier; a large (0.8 km long) landslide which encompasses native slope terrain and a portion of the northern spoil piles; Leviathan Creek which is in a concrete lined channel through the southern portion of the mine spoil; and Aspen Creek, which flows along the eastern edge of the landslide and discharges into Leviathan Creek. The mine spoil is subdivided on Figure 2 with a 32-hectare area directly north of the pit hereafter referred to as the northern spoil area.

Elevations at the mine site range from 2,010 to 2,320 meters above sea level. Leviathan Creek has a steep gradient within the mine area, dropping 137 vertical meters in less than 1.6 kilometres. Leviathan Creek lies within the Bryant Creek Basin, comprising a 90 square kilometre area within the Carson River catchment. The area receives approximately 510 millimetres of precipitation a year, primarily from winter snowstorms.

The northern spoil area and the area immediately downslope to the northwest are the focus of this paper. This area consists of terraced or benched spoil piles up to 30 meters in thickness. The spoil consists of altered and mineralized overburden that was removed from the pit area and subsequently bottom or end dumped along the slope to the north of the pit. The topography slopes to the north and northwest towards Leviathan Creek (Figure 2). The northern spoil area is bounded on the northeast by Aspen Creek. The large landslide north of the pit is a historical feature. This slope failure is essentially a block slide with some slumping occurring in the portion of the slide comprising the spoil. The overall relief along the landslide from the crown within the spoil to the Leviathan Creek channel is approximately 180 meters.

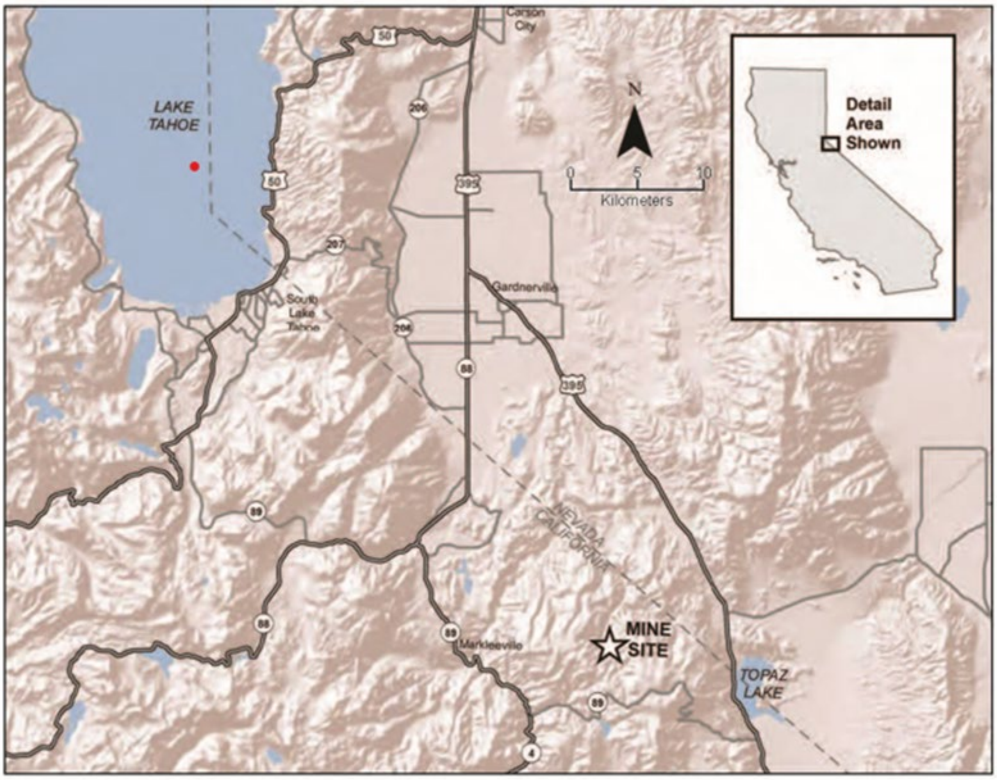


Figure 1 Leviathan Mine location map

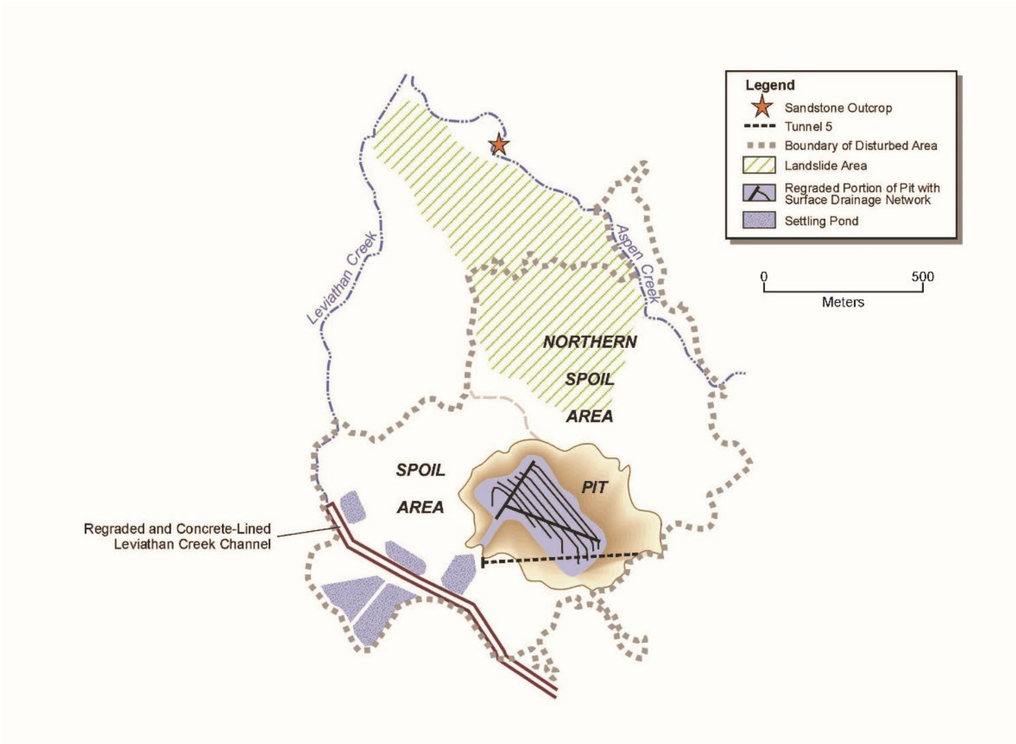


Figure 2 Current Leviathan Mine features

2 Hydrogeologic background

The mine is located in a northeast trending belt of mines and mineralized areas comprising the Silver Mountain Mining District and the Monitor Mogul Mining District. The mining belt is in a regional Tertiary Period volcanic field within the Walker Lane structural belt (Walker Lane) and the central Sierra Nevada – Basin and Range transition. The mines and mineral deposits in the belt are part of the Western Andesite magmatic assemblage described by John (2001). The assemblage is characterized by epithermal (i.e., shallow hydrothermal) gold-silver deposits within the Walker Lane.

The Leviathan Mine has been identified as a high-sulfidation epithermal deposit by McKnight & Vikre (2022). The sulfur ore body was deposited as part of a volcanic-hydrothermal vent system with corresponding hydrothermal alteration and mineralization of several host rock lithologies of the Relief Peak Formation. The Relief Peak Formation (Slemmons 1966) is Miocene in age and forms the basal volcanic layer across the region and the mine site. It is a complex assemblage of andesitic flows, flow breccias, shallow intrusions, and minor basalt flows, along with volcaniclastic (tuffaceous) sandstones and lahars. The stratigraphy of unaltered Relief Peak country rock near the ore body at the mine consists of a lower sedimentary sequence of tuffaceous sandstones and lahars, predominantly of andesitic composition. This lower sequence is overlain by an intervening middle basalt flow. Overlying the basalt flow is an upper sedimentary sequence of tuffaceous sandstone and lahars, also predominantly of andesitic composition.

The initially hot and highly acidic fluids from the hydrothermal vent system leached the country rock, thereby altering the rock to clay minerals and alunite and leaving an advanced argillically altered former upper sedimentary sequence. Basalt near the location of the ore deposit was also leached. The altered rock units are weak and friable due to the acidic leaching from the alteration. The acidic leaching left silica from the country rock at or near the surface to form silica cap (silicified caprock) deposits, also known as the quartz pyrite jasperoid of John et al. (1981).

Following the silicification hydrothermal pulse, hydrogen sulfide-charged and iron-copper-bearing fluids moved upward (through fractures), and sulfur was deposited from oxidation of the hydrogen sulfide gas.

Essentially, the tuffaceous sandstone became a sink for the sulfur. Later stages of sulfur deposition infilled fractures and channels in the ore body and infilled some fractures in the underlying basalt. Pyrite was deposited near and within the sulfur ore body from iron-bearing fluids under oxygen-poor conditions. Pyrite occurs pervasively in the altered upper sedimentary unit within the pit area and is consequently distributed throughout the spoil.

Groundwater recharge at the mine occurs predominantly by infiltration from snowmelt, including on the disturbed portions of the Site (SSPA 2022). Groundwater flow essentially follows topography and occurs primarily within the mine spoil, underlying native materials, and weathered bedrock. Groundwater flow at the pit and west of the pit is generally to the south and southwest toward Leviathan Creek. Groundwater flow north of the pit is toward the north to northwest, including within the landslide. Bedrock underlies the mine spoil and the native unconsolidated materials and is comprised of a variety of volcaniclastic sedimentary units and extrusive volcanic rock types. Generally, the upper portion of the bedrock is weathered and is hydraulically connected to the overlying unconsolidated materials. The weathered bedrock and overlying unconsolidated materials are designated the unconsolidated (hydrogeologic) unit. Overall, the hydraulic conductivity of the unweathered bedrock is significantly less than that of the overlying mine spoil and unconsolidated materials, except where significantly altered near the pit and in some sedimentary units. Consequently, groundwater flow in bedrock is minor relative to the unconsolidated unit, but potential downgradient migration of mine affected groundwater in preferential pathways, such as fracture zones in the bedrock, is of concern to the regulatory agencies.

Acid rock drainage occurs at the mine and is present in groundwater and surface water. The ARD is characterized by high sulfate, low pH, and high concentrations of certain metalloids and metals such as arsenic and nickel, respectively. The oxidation of pyrite and, to a lesser extent arsenopyrite and sulfur, within hydrothermally altered ore rock and mine spoil, is the source of ARD. Ball & Nordstrom (1985) linked the ARD at the mine predominantly to the oxidation of pyrite. The pyrite at the mine is highly reactive and oxidizes rapidly; in part due to the fine-grained to cryptocrystalline occurrence of the pyrite (Pabst 1940). Overall, this increases the potential for generation of ARD at the mine.

3 Calcite occurrence at Leviathan

Calcite is commonly found with epithermal deposits, including those within Walker Lane, occurring in mineral assemblages associated with propylitic or argillic alteration as described by Simmons et al. 2005 and Simmons & Christenson 1994. Calcite associated with propylitic alteration forms as replacements of rock-forming minerals at relatively high temperatures (i.e., generally greater than 240° C) deep within the epithermal environment. Calcite associated with argillic alteration forms as platy crystals filling voids or fractures at cooler temperatures (i.e., generally less than 180°C) on the periphery of shallow epithermal environments; generally occurring at a restricted vertical interval of a few hundred meters. The calcite deposits form through alteration by steam-heated waters rich in carbon dioxide (CO_2) (Simmons *et al.* 2005). It precipitates through exsolution of CO_2 in boiling fluids from the sulfidation system. The presence or absence of calcite in the mineral assemblage is directly related to the concentration of CO_2 in the fluid.

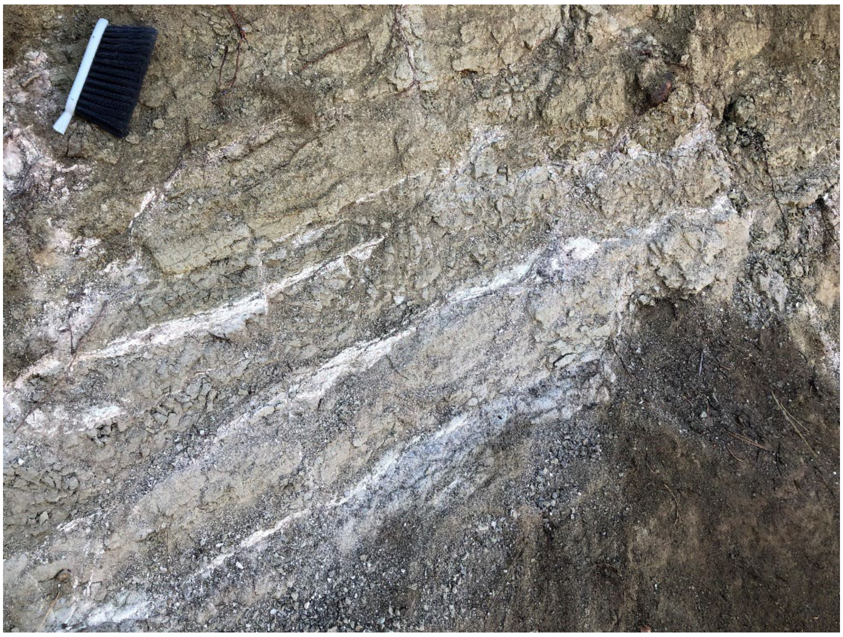
The presence of calcite at the Leviathan Mine has been investigated as components of several geologic and hydrogeologic studies conducted during the past 50 years. Geologic studies by Herbst & Sciacca (1982) and Sciacca (1984) were the first to note the presence of calcite in the native materials, and the volcaniclastic sandstones and lahars downslope and underlying the northern portion of the mine spoil. Subsequently, the U.S. Geological Survey (USGS) advanced several boreholes and documented the occurrence of calcite from x-ray diffraction (XRD) analysis of rock samples (Hammermeister & Walmsley, 1985). In the 2010's, as part of a Remedial Investigation (RI) conducted at the Leviathan Mine, 23 boreholes were advanced in, surrounding, and downslope of the northern spoil piles (WSP USA, 2023, in review). Detailed logs were prepared from the well borehole drill cores. This included systematic checking for the presence of calcite in the core. The calcite occurrences were noted and recorded on the borehole logs. In addition, a sandstone outcrop containing calcite in fractures north of the landslide, described in Sciacca (2008) and shown on Figure 1, was cleared of slope wash to view and document the occurrence of calcite within the sandstone outcrop. The exposure revealed the pervasive occurrence of calcite filling low angle bedding plane fractures as well as high angle fractures within the volcaniclastic sandstone (Figure 3). Higher in the section the calcite occurrence was less prevalent where bedding plane fracture filling was not evident, though high angle fracture filling was.

In the USGS study, calcite was found to be present within the sedimentary unit underlying and downslope of the northern spoil pile. This included four of five boreholes from which samples were collected and nine of 13 samples of the sedimentary unit that were submitted for XRD analysis. The XRD analysis showed no samples of mine spoil or hydrothermally altered material contained calcite. Samples were selected for evaluation of general mineralogy and clay type to support a study of the landslide; and were not selected based upon a systematic scheme to characterize the occurrence of calcite.

Based upon the detailed logging for the RI and previous characterization efforts, calcite was confirmed in 25 boreholes surrounding the pit. Calcite is present within unaltered volcaniclastic and volcanic rock, and not within caprock, argillically altered units, or ore rock. S.S. Papadopulos & Associates Inc. (SSPA, 2022, pp. 16 through 18) provided a detailed account of the logged occurrence of calcite in these 25 boreholes. Calcite is present in rock units surrounding the hydrothermally altered rock in the pit to the north, west, and south. This includes unaltered rock units under and downslope (downgradient) of the northern spoil piles.

In summary, the studies that have been conducted have shown that calcite at the Leviathan Mine is present as a secondary mineral within the unaltered sedimentary units, filling joints faces in unaltered andesite, as a fracture and vug filling cement within unaltered sandstone and lahars of the lower sedimentary unit to the north of the pit, and as root coatings in soil derived from unaltered sedimentary units (Herbst & Sciacca 1982; Sciacca (1984). At the sandstone outcrop, the exposure revealed the pervasive occurrence of calcite filling low angle bedding plane fractures as well as high angle fractures within the volcaniclastic sandstone (Figure 3). In addition, the studies have shown that calcite has not been observed in any hydrothermally altered units at the mine. Thus, the calcite was deposited away from the likely hydrothermal vent location at the centre of the pit area. The presence of calcite around the periphery of the argillically altered units at the mine and its occurrence as platy crystals or crust filling fractures and voids fits the previously described second model for calcite deposition in a shallow epithermal system. The occurrence and areal distribution of the calcite fit well with the epithermal (CO_2) outgassing model described in the literature (Simmons et al. 2005; Simmons & Christenson 1994).

(a)



(b)

Figure 3 Photographs of sandstone outcrop. (a) Exposure of sandstone with calcite filling fractures; (b) Closeup of outcrop in upper photo showing pervasive filling of calcite in fractures; brush is 17 cm in length

4 Groundwater quality and evidence of buffering

An extensive groundwater monitoring system, consisting of monitoring wells, piezometers, and seeps, was sampled as part of the RI for the Leviathan Mine site. The strategically selected well, piezometer, and seep samples were analysed for a comprehensive suite of constituents following US Environmental Protection Agency (EPA) protocols. These included field pH, dissolved metals, general chemistry parameters, sulfate, and bicarbonate. Based on these data, the extent of acidic mine-influenced groundwater (groundwater with a pH of less than 6.5) was defined in the unconsolidated unit (Figure 4) and the bedrock (Figure 5). The analytical results were compared to the occurrence and distribution of calcite at and downslope of the northern spoil piles to evaluate the extent of buffering of acidic-mine influenced groundwater by calcite. The wells in which calcite was noted within the well borehole are noted on Figure 4 and Figure 5.

In the Unconsolidated Unit (Figure 4**),** the lowest pH values were from wells and seeps within the spoil piles with wells MW-39 and MW-31, both having values of 2.8. Only two well boreholes in the unconsolidated unit had calcite in the completion intervals. Well MW-43 had calcite present and a pH of 7.8 which restricts the southwestern extent of ARD downgradient near the toe of the landslide. Seep SD-08 had a pH value of 7.7 and currently restricts the north-eastward extent of ARD in the toe of the landslide.

In the bedrock (Figure 5), acidic mine-influenced groundwater is restricted to under the southernmost portion of the northern spoil piles, in the vicinity of MW-28. MW-28 is completed in pyrite-bearing hydrothermally altered rock. The remaining wells under the northern spoil piles and downslope (downgradient) area extending to Leviathan Creek all had calcite present within their completion intervals and as such, had groundwater pH values above 6.5. The presence of calcite in all bedrock wells that had circumneutral or alkaline pH values is evidence of the potential for buffering from calcite if acidic mine-influenced groundwater were to migrate to these locations.

Evaluation of pH, sulfate, and bicarbonate in combination in select wells provides evidence of buffering by calcite in the northern area. Bicarbonate is a product of a buffering reaction between calcite and sulfuric acid in the ARD. Sulfate is less affected by the buffering reactions and therefore sulfate concentrations are minimally affected by calcite buffering. Two sets of monitoring locations provide evidence of the buffering that has occurred: monitoring wells MW-24 and MW-26 and seeps SD-10 and SD-08.

MW-26 is a monitoring well completed in the mine spoil and MW-24 is a bedrock well at the same location. There is a downward hydraulic gradient, and sulfate concentrations are similar in both wells indicating that there has been downward migration of groundwater from the mine spoil into the bedrock (Table 1). The pH value in MW-26 is 3.4 whereas it is 7.0 in MW-24 and bicarbonate concentrations are <2 mg/L and 141 mg/L, respectively, indicating that calcite buffering has occurred.

Table 1 Chemical concentrations at selected locations

Location	pH	Sulfate (mg/L)	Bicarbonate (mg/L)
MW-24	7.0	1280	141
MW-26	3.4	1638	<2
SD-08	7.7	1010	135
SD-10	5.5	1180	<2

Note: Average concentrations from samples collected in 2016 and 2017.

SD-10 and SD-08 are seeps on the lower portion of the landslide. Sulfate concentrations are similar in each, 1180 mg/L and 1010 mg/L, respectively. The pH value in SD-10 is 5.5 whereas it is 7.7 in SD-08 and bicarbonate concentrations are <2 mg/L and 135 mg/L, respectively. This indicates that calcite buffering has likely occurred, and is likely still occurring, on the lower portion of the landslide.

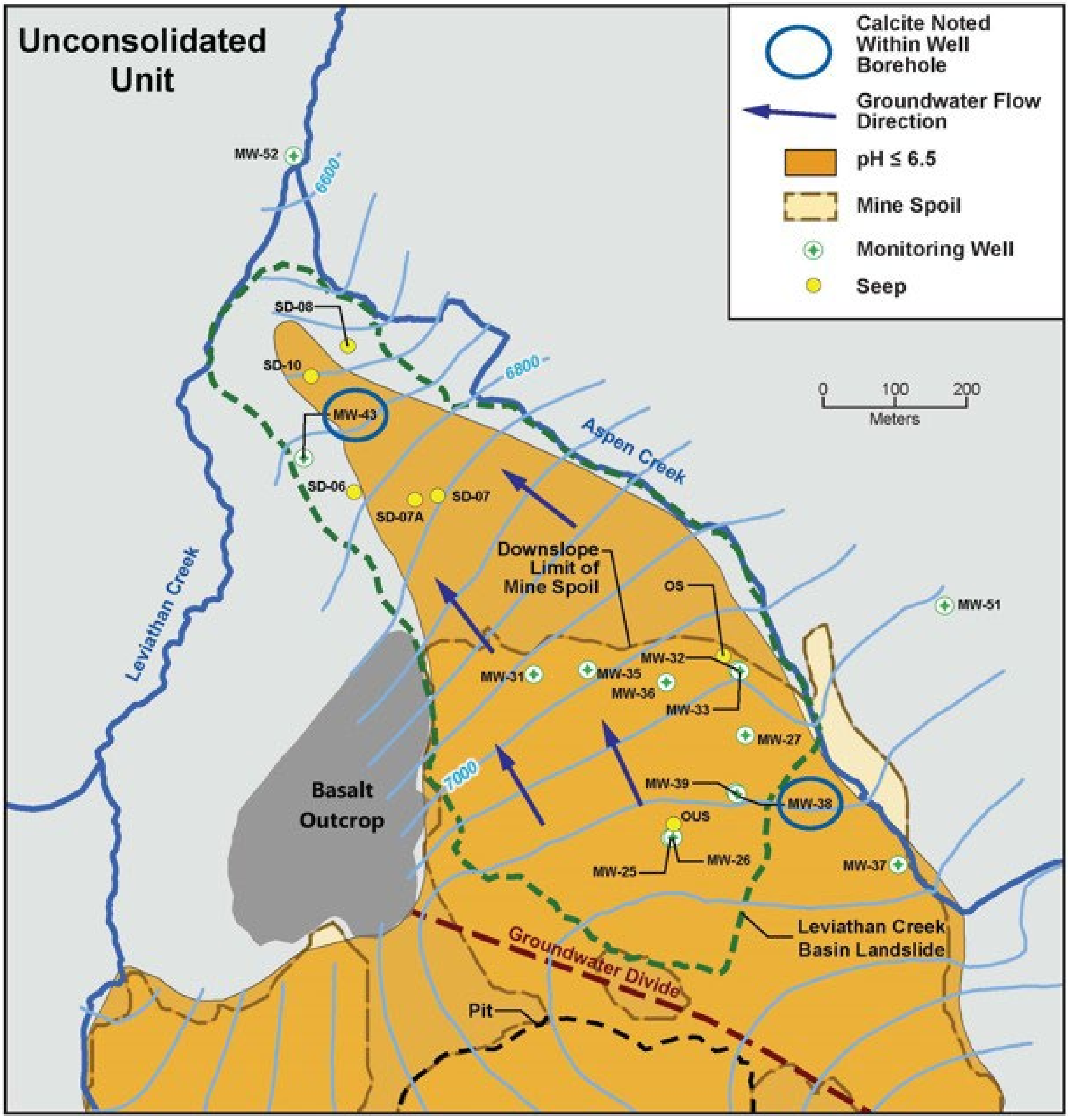


Figure 4 Unconsolidated unit groundwater map with calcite occurrence

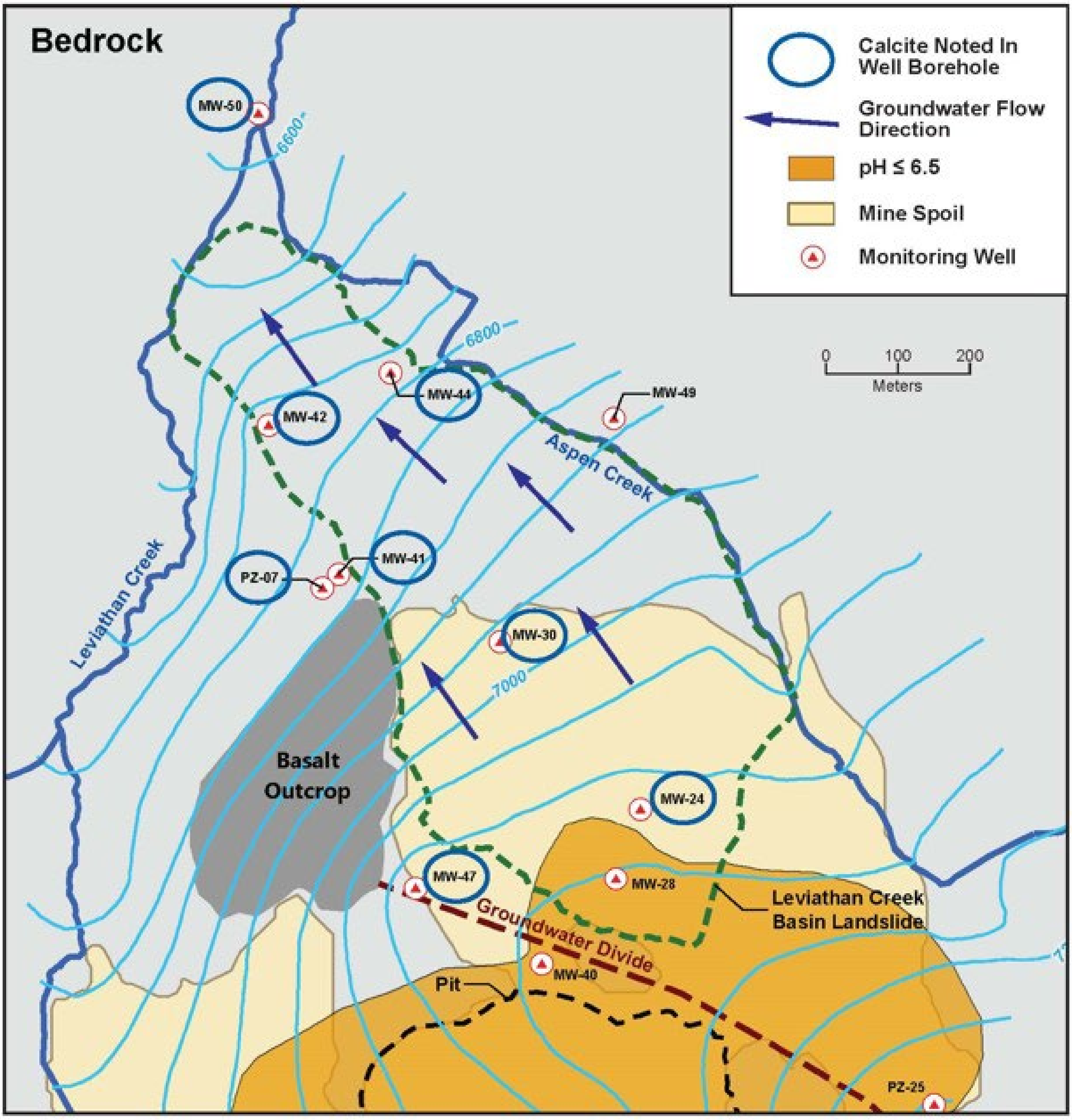


Figure 5 Bedrock groundwater map with calcite occurrence

5 Summary and conclusions

Calcite is present within unaltered sedimentary (volcaniclastic) and volcanic rocks surrounding the open pit of the mine; the likely hydrothermal vent location of the epithermal mineral deposit. The calcite predominantly fills fractures and voids within these rocks and can exhibit pervasive occurrence.

Groundwater within the area north of the pit occurs within the northern mine spoil and shallow material comprising the unconsolidated unit. It also occurs within the deeper bedrock and both units have groundwater flow downslope to the northwest towards Leviathan Creek. Within the unconsolidated unit ARD is present within the northern spoil pile and extends downgradient (northwest) towards Leviathan Creek. Calcite is limited in extent within this horizon but, where present, may restrict the lateral extent of ARD at the toe of the landslide and vertically at one location at the northern spoil piles.

ARD within bedrock groundwater is restricted to a small area under the southern portion of the northern overburden piles. This coincides with the large areal distribution of calcite within bedrock wells and supports the buffering of ARD by calcite. An additional line of evidence is the prevalence of bicarbonate (a reaction product of the buffering reaction between calcite and sulfuric acid from ARD) and higher pH values in wells with calcite present within their completion intervals. These multiple lines of evidence support the theory of buffering of ARD by naturally occurring calcite. This and other potential natural attenuation mechanisms should be evaluated for sites where ARD may be or is present, and the natural geological conditions avail themselves to such a solution.

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