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Mine water composition in abandoned copper mines of the UNESCO Global Geopark Ore of the Alps

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Abstract

The central theme of the UNESCO Global Geopark “Ore of the Alps” in Salzburg’s Pongau region is prehistoric and historic copper mining in the Eastern Alpine Greywacke Zone. Much is known about the history and geology of the ore deposits. However, to date the effects of mining on groundwater quality is not investigated yet. From January to July 2024, monthly samples were taken from mine water outlets and analysed for field parameters, main and trace elements as well as sulphate isotopes. The quality of mine water was compared with reference stations sampled at the same time. The measurements show that the pH values of the mine water are in the slightly alkaline range, meaning that the heavy metal concentration is mostly harmless, although the mine water has significantly higher concentration than the reference water. The sulphate levels in the mine waters are significantly higher than those of the reference waters. Isotope analyses indicate that the increased sulphate levels are due to sulphide oxidation and thus to mining. Although systematic discharge measurements were not possible observations in one of the mines indicate a positive correlation between discharge and mineralisation. The study shows that historical mining continues to affect water composition. However, as the main ore is chalcopyrite and the mineralisation of the ore veins is bound to carbonates, the effects of sulphide weathering in the aquifer are buffered. The mine water remains pH-neutral to slightly alkaline, so it has only a low potential to dissolve harmful heavy metals. The study provides an overview of the current groundwater situation in the ‘Ore of the Alps’ UNESCO Global Geopark and confirms that there is no groundwater contamination risk from historic mining facilities. It thus contributes to a holistic understanding of this specific geo-heritage.

1. Introduction

Water drainage and the associated treatment of groundwater affected by a mine (by definition mine water (Burghardt et al., 2017) is an important issue in almost all mining operations worldwide. This is true for active as well as for abandoned mines. Questions about mine water quality are an essential part of post-mining research, as evidenced by numerous studies (Kessler et al., 2020 and citations therein). It is well known that water quality might be influenced for decades after mining operation and can cause substantial environmental problems (Sengupta, 2021; Wolkersdorfer, 2008). Depending on the mineralogical composition of the ore and the host rock acid mine water or circumneutral to alkaline water emerges from mines. Acid mine waters are produced by the contact of water and oxygen with sulphide when there is an insufficient amount of buffering minerals available Nordstrom, 2011). Circumneutral or alkaline mine waters occur where buffering minerals dominate the hydrochemical processes (Younger and Robins, 2002). However, it can be observed that mine water quality usually improves over time after the first flush (Younger, 1997) which is dominated by leaching of secondary minerals and salts (Merrit and Power, 2022; Mugova et al., 2024).

From a hydrogeological perspective, adits form underground flow paths that are comparable to fractured or karst aquifers in terms of the prevailing flow dynamics, depending on the type of mining (Bedoya-Gonzalez et al., 2023; Li et al., 2013). Thus, mines are artificially created cavities that influence the natural flow conditions in the aquifer (Bucher et al., 2012; Behzad et al., 2022; Golian et al., 2021). New flow paths are created, water residence times in the rock mass are altered, and mining voids allow meteoric freshwater to flow into areas that would be free of water and oxygen without human influence.

In particular, the addition of oxygen to sulphide ores leads to chemical reactions that affect mine water, as illustrated by the examples of pyrite and chalcopyrite weathering in formulae 1 and 2, which both lead to an enrichment of sulphate:

$$FeS_2 + H_2O + 3.5\ O_2 \rightarrow Fe^{2+} + 2SO_4^{\ 2-} + 2H^+ \qquad \text{Oxidation of Pyrite} \quad [1]$$

$$CuFeS_2 + 4O_2 \rightarrow Cu^{2+} + Fe^{2+} + 2SO_4^{2-} \qquad \text{Oxidation of Chalcopyrite} \quad [2]$$

While pyrite weathering occurs relatively quickly and leads to acidification, chalcopyrite weathering is slower and does not directly lead to acidification. However, chalcopyrite weathering is accelerated by prior pyrite weathering and both processes cause copper, iron, and sulphate to dissolve in the water.

Released Fe^{2+} is then oxidized to Fe^{3+} according to Formula 3:

$$Fe^{2+} + O_2 + H^+ \rightarrow Fe^{3+} + H_2O \qquad \text{Oxidation of } Fe^{2+} \quad [3]$$

Depending on the surrounding environment and the availability of other reaction partners, sulphide oxidation can be followed by acidification and/or by the formation of secondary minerals. Examples of possible precipitation reactions are shown in formulae 4 and 5.

$$2Cu^{2+} + CO_3^{2-} + 2OH^- \rightarrow Cu_2CO_3(OH)_2 \qquad \text{Malachite} \quad [4]$$

$$Fe^{3+} + 3H_2O \rightarrow Fe(OH)_3 + 3H^+ \qquad \text{Iron hydroxide} \quad [5]$$

Low pH values promote the dissolution of metals from the surrounding rocks, so that emerging mine waters are often not only acidic and high in sulphate, but are also enriched in various toxic metals and metalloids such as arsenic (Veskovic´ et al., 2024). On the other hand, the formation of secondary minerals causes dissolved ions to be bound. Heavy metals can also be adsorbed onto secondary minerals. The composition of mine water at a sampling point reflects all these processes, and a precise analysis of a variety of ion ratios can help to reconstruct the history of the water.

However, the question of whether elevated sulphate concentration is due to evaporite dissolution or sulphide oxidation as a typical process associated with mine waters can only be conclusively answered using sulphate isotopes. In the sulphate molecule (SO_4^{2-}), different isotopes can occur for both sulphur and oxygen. The most common isotope of sulphur is ^{32}S and that of oxygen is ^{16}O. Additionally heavy isotopes ^{34}S and ^{18}O occur, each with two extra neutrons in the nucleus. Isotopes of the same element have the same chemical but different physical properties. As a result, the ratio of light to heavy isotopes changes during physical or biochemical processes. This fact can be used to determine the origin of sulphates in mine waters (Rinder et al., 2020). The ^{18}O values in the sulphate molecule originate either from evaporitic sediments and therefore vary depending on the formation period and conditions, or from recent groundwater and atmosphere that is involved in the dissolution of sulphidic ores. The ^{34}S values, on the other hand, originate either from the evaporites or the sulphides. In general, positive δ-values for ^{34}S are typical of Mesozoic evaporites while sulphides are characterized by negative δ-values. Thus, the use of sulphate isotopes can provide additional evidence of the ongoing influence of mining on water composition.

For many mines that have been closed decades ago or longer, no post-mining water treatment measures were taken (Nascimento et al., 2023). The mine water was, and in many cases still is, discharged into the receiving streams in an uncontrolled manner. Several old mining sites are being studied to determine how the mine water quality evolves after decades without human intervention. While some studies show that natural attenuation through leaching in the mines, adsorption or precipitation and dilution by rainwater can lead to an improvement in water quality over the decades (Cidu et al., 2011; Yousefi, 2022), at other locations no significant improvements in water quality have been observed in the receiving streams (Nascimento et al., 2023; Gilchrist et al., 2009).

The Eastern Alps and the Greywacke Zone in particular, are known for significant ore deposits and the associated mining history (Breitenlechner et al., 2014; Pernicka et al., 2016, GeoSphere Austria, 2025b. The “Ore of the Alps” UNESCO Global Geopark (UGG) in the Austrian district of Pongau in the province of Salzburg is dedicated to prehistoric and historic copper mining. Exhibition mines in Mühlbach, St. Veit and Hüttau, inform visitors about the copper mining that took place in the region for centuries (Erz der Alpen UNESCO Global Geopark, 2024). A study of mining and smelting sites in the province of Salzburg (Feitzinger et al., 1998) also includes the sites in the Ore of the Alps UGG. Mining geology of the deposits was described, ore mining in the region was examined from a historical perspective, and the metallurgical sites

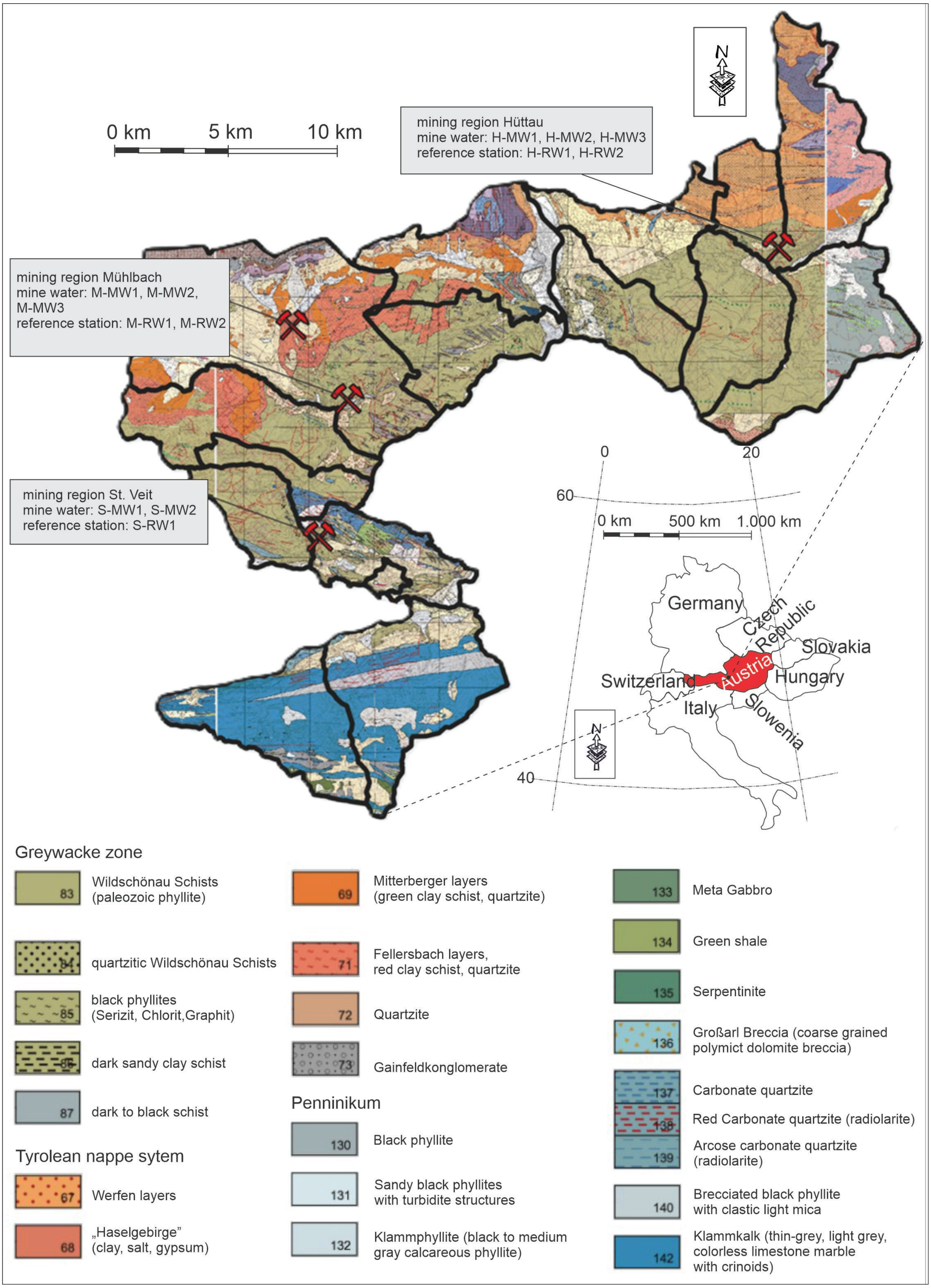

Figure 1: Overview of the study region with the three mines and their geological setting. Geology based on (Pestal et al., 2009a; GeoSphere Austria, 2025a).

were presented from an environmental protection perspective. Soil contamination from tailings and flue gas emissions were discussed. However, mine water has not been systematically recorded or assessed. This may indicate that the mine water currently discharged from old mines does not represent a significant environmental hazard, otherwise it would have been detected over the past decades. Considering the general processes of sulphide weathering described in formulae 1 and 2, the question arises as to why this is the case. Is it because the contaminants have been leached out of the mine over a sufficiently long period of time, i.e. natural remediation is complete? Are special geological conditions in the mining area responsible for the fact that sulphide weathering and the associated release of metals and metalloids do not take place at all, or has existing mine water contamination simply not been recognized until now?

In any case, the influence of underground mines and dumps on the composition of mine waters in the prehistoric and historic mining areas of the Ore of the Alps UGG is relevant for a holistic understanding of this specific mining region. Furthermore, the results can contribute to an understanding of the general processes of natural remediation and their controlling factors and thus can be transferred to other historic and prehistoric mining regions in comparable geological settings.

2. Study area

The Ore of the Alps UGG is located in the northern part of the district of Pongau in the Austrian province of Salzburg along the tributaries to the Salzach Valley. The Geopark covers an area of 211 km^2 in the four municipalities of Hüttau, Bischofshofen, Mühlbach am Hochkönig and St. Veit im Pongau. It is a region where mining has been practiced since the second millennium BC (Breitenlechner et al., 2014; Pernicka et al., 2016), especially copper has been mined here until the last active mine in the region was finally closed in 1977 (Feistzinger et al., 1998). The climate of the study area is humid, with a mean annual precipitation of about 1200 mm and mean annual temperatures around 7 °C (Austrian Hydrographic Service, 2024). The highest temperatures are around 20 °C between June and August and lowest temperatures around -4 °C between December and March. There is usually a snow cover from November to April, the average snow cover thickness is about 2 m (Austrian Hydrographic Service, 2024). The surface water system in the area is based on the Salzach River, which drains the entire area from the Central Gneiss region of the Hohe Tauern in the south to the Alpine foothills in the north.

The copper deposits are located on the northern margin of the Eastern Alpine Greywacke Zone, near the base of the Northern Calcareous Alps. In the former mining area, an ancient Paleozoic sequence of sericite phyllites and quartzites which are locally intercalated with black phyllites, green schists and diabase represents the dominant lithology. Small-scale limestone marbles and limestone schists in the form of elongated rock structures are described as intercalations in the siliciclastic metasediments (Pestal et al., 2009b). The rocks of the Greywacke Zone are overlain in higher elevations of the respective catchments by Werfen layers with sandstones and quartzites, partly sandy clay shales and, to a lesser extent, conglomerates (Pestal et al., 2009b). The general layer dip of the strata is north-trending, with local folds and dips showing other orientations (Pestal et al., 2009a). Based on the predominant lithologies (schist, phyllite, diabase), all the areas investigated can be classified as low-conductivity fractured aquifers (Schubert, 2006). Even the overlying Werferner units (here mainly represented by sandstones and quartzites) are more likely to be classified as aquitards and despite relatively high annual precipitation groundwater recharge is supposed to be low. The artificial water pathways of the mining adits thus form drainage systems through which precipitation water can infiltrate faster but also be drained off more quickly.

The mines investigated are vein deposits that are either unconformably cut through the host rock or concordantly intercalated into the schistosity. In each mine chalcopyrite mineralization is dominant. In some intervals, mineralization has been characterized by elevated nickel content in the form of gersdorffite (NiAsS) (Pernicka et al., 2016). Other ores are associated with the deposits (e.g. pyrite, magnetopyrites, arsenopyrite). Typical ore veins are embedded in carbonates of the magnesite-siderite and dolomite-ankerite series (Feitzinger et al., 1998).

There are numerous old mine in the region. However, most of them are no longer accessible, and/or there is no reliable information about their extent. For the present study three accessible adit systems were selected: (1) Mitterberg in Mühlbach, a mine in Hüttau, in the Larzenbach area, and another in St. Veit, Sunnpau were investigated (Fig. 1). The investigated mines are located between 900 and 1200 m a.s.l., the catchment areas range in altitude from 1800 m a.s.l. in Hüttau to over 2400 m a.s.l. in the Mitterberg area.

In the Mitterberg mining district the mineralization is hosted by sericite phyllites, which are intercalated with quartzites, black phyllites, green schists and diabase (Feitzinger et al., 1998).

The mine in Hüttau is located on the orographic right flank of the Larzenbach trench, 600 m north of the village of Hüttau at an elevation of 700 - 800 m above sea level. Ore veins were developed in Wildschönauer Schiefer (paleozoic phyllitic schists which are the dominant unit in the Eastern Alpine Greywacke Zone). The mine was converted to an exhibition mine in the 1990s after being closed for over 100 years. It is now accessible from two levels (Georgstollen and Barbarastollen).

The Sunnpau Mine in St. Veit is characterized by dolomitic calcareous phyllites. The fahlores occurring here are bound to Silurian-Devonian limestones and dolomites (Feitzinger et al., 1998). The mine was closed in the late 19th century and is now operated as an exhibition mine.

Figure 2: Photo panel showing the mine water measuring points in the three mines. **(a)** M-MW-1 and **(b)** M-MW2 represent the adits of Rupertistollen and Emilstollen in Mühlbach. **(c)** H-MW1, **(d)** H-MW2, and **(e)** H-MW3 are small ponds in the adits that are fed from below. **(f)** S-MW1 is discharge from the tunnel roof, and **(g)** S-MW2 is a pond with inflow from below.

3. Methods

3.1. Selection of sampling stations

Representative points for repeated sampling of mine water and groundwater were identified during site and adit inspections. The aim was to select samples representing the following types of water: (1) water that has clearly circulated within the mines, (2) water that may be influenced by mining dumps, (3) water that circulates outside the influence of known mining areas but in a comparable geological-hydrogeological environment and can therefore serve as a reference. The identification is based on information provided by the mining management, plans of the adit systems, searches in the water database of the Environment Department of the Provincial Government of Salzburg, literature searches and own field work. As a result of this search, seven mine water outlets and four reference springs for monthly sampling were identified. No water outlets from mining waste dumps could be located in the respective regions.

3.2. Sampling, hydrochemistry and isotope analysis

The selected monitoring sites were sampled six times over a seven-month period (January to July 2024). 0.7 L of filtered water was collected in PE bottles (rinsed three times with sample water) (0.1 L for cation analysis, filtered with 0.45 µm Millipore filter, acidified with 1M HNO_3 to pH < 2, 0.1 L for anion analysis, filtered with 0.45 µm Millipore filter, 0.5 L for hydrogencarbonate (HCO_3) determination and as reserve sample). All bottles were filled to the brim to prevent air bubbles. Samples were kept refrigerated until analysis. The field parameters specific electrical conductivity, pH value, and temperature were measured on site with a multiparameter measuring device WTW Multi 350i with sensors TetraCon 325 and pH Electrode Sentix 41. The measurement of redox potential and oxygen was omitted, as these values are expected to be significantly influenced by the respective atmosphere at the available measuring points (adits, ponds in the mines). The water was analyzed in the hydrogeology laboratory of the University of Salzburg. Anions (Cl^-, SO_4^{2-}, F^-, Br^-, and NO_3^{2-}) and major cations (Ca^{2+}, Mg^{2+}, Na^+, and K^+) were analysed by ion chromatography (Dionex Aquion, Thermo Fisher), minor cations by ICP-MS (Agilent 7.900). Hydrogencarbonate was determined titrimetrically (Titroline 5000, SI Analytics) not later than one day after sampling. In each case, titration was carried out up to a pH value of 4.3. Individual samples had a pH value > vb8.2. However, in these cases, a separate determination of the CO_3 content by titration to pH 8.2 was not carried out; instead, the titration result was converted into HCO_3 concentration.

Additional samples (1L) were taken once for sulphate isotope analysis. The analysis of $\delta^{34}S$ values in sulphate was carried out by Imprint analytics GmbH (Neutal, Austria) using a Eurovector elemental analyser coupled with a NU Horizon IRMS after the method described in Révész et al. (2012). $\delta^{18}O$ values were determined by the same lab with a Hekatech TCEA coupled to the NU Horizon IRMS. The isotopic analyses of sulphur and oxygen in sulphate are reported in the δ-notation relative to a standard (VCDT and VSMOW, respectively). The stable isotope ratios represent the mean value of the repeat analyses of a sample ($n \geq 2$). The values were accepted if the standard deviation of the repeat analysis was ≤ 0.8 for $\delta^{18}O$ and ≤ 1.0 for $\delta^{34}S$. Sulphate-enriched mine water samples from the April 2024 sampling campaign were analysed. Data interpretation, ion balance calculations and visualization were performed using AquaChem 12.0 software (Waterloo Hydrogeologic, 2022).

4. Results

In the areas of the three mines, mine water discharge, and reference springs clearly unaffected by mining were sought. Figure 1 shows the locations of the monitoring sites in the municipalities of Mühlbach, Hüttau and St. Veit. Figure 2 shows mine water sampling sites. The stations were labeled as MW for mine water and RW for references water combined with abbreviations for the respective mining region (H = Hüttau, M = Mühlbach, S = St. Veit). Table 1 presents the results of six sampling campaigns as median values for physicochemical parameters. The complete data set is available as Supplement 1. According to the postulated low hydraulic conductivities in the dominant phyllite lithologies little water inflow was detected in all three mining regions. A total of seven mine water outlets and four reference stations (springs and streams) were sampled. In the Mitterberg area, all former mines have either collapsed or were walled up when the mine was closed in 1977. However, the entire drainage of the adit systems is accessible for sampling at the exits of Rupertistollen, (M-MW1) and Emilstollen (M-MW2) In addition, the Schrammbach stream (M-RW2), upstream of the tunnel and a nearby spring (M-RW1), also upstream of the adits, were sampled for reference. In Hüttau, three different springs in the lower Georgstollen were selected for sampling (H-MW1, H-MW2, H-MW3). A nearby unnamed stream (H-RW1) was used as a reference station for this adit system, since no suitable springs could be found in the vicinity. In the so-called Mathiasstollen in St. Veit, a water discharge from a fracture (S-MW1) and a small pool (S-MW2) were selected. The pool is formed by water entering below the water table and can only flow out through the adit system when it reaches a certain level. For reference, the nearby Harbergbach (S-RW1) was sampled at a point above the mine.

In addition to the self-sampled water quality data, hydrochemical data of mine water discharge from the Emilstollen (M-MW2e) and Barbarastollen (M-MW3e) (e for external data) were provided by the Austrian Geological Survey (Schubert et al., 2022). Unpublished data from M-MW2, sampled in the year 2020 were provided by the Environment Department of the Salzburg Provincial Government. This additional data is included in the results presented (Tab.1, Suppl. 1, and Figs. 4–7).

mining region		Mühlbach					St. Veit			Hüttau				Reference	Mine water	ratio mine/ref.
sampling station		M-RW1 (n=6)	M-RW2 (n=6)	M-MW1 (n=6)	M-MW2 (n=11)	M-MW3 (n=1)	S-RW1 (n=6)	S-MW1 (n=5)	S-MW2 (n=6)	H-RW1 (n=5)	H-MW1 (n=5)	H-MW2 (n=6)	H-MW3 (n=6)	n=23	n=46	
parameter	unit															
pH		7.9	8.4	8.2	7.8	7.2	8.6	8.3	8.6	8.4	8.5	8.3	8.4	8.4	8.3	0.99
Temp	°C	8.0	4.9	7.3	8.3	7.2	7.9	4.8	5.9	5.3	8.4	6.7	5.4	6.6	7.2	1.08
Cond	µS/cm	316.5	284.0	428.0	948.0	349.0	332.0	277.0	512.0	277.0	840.0	545.0	499.5	308.5	514.0	**1.67**
K	mg/L	1.4	1.2	1.2	1.2	0.3	1.6	2.7	1.1	1.6	7.6	2.2	2.0	1.4	1.5	1.10
Li	mg/L	0.6	0.6	0.6	0.5	0.0	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.97
Na	mg/L	4.8	5.4	2.5	12.5	5.5	2.7	2.4	1.9	3.9	89.2	7.6	11.8	3.9	7.6	**1.96**
Mg	mg/L	21.6	17.2	19.9	46.8	12.8	11.0	6.6	51.9	12.7	39.8	51.6	34.6	14.4	39.0	**2.70**
Ca	mg/L	33.2	29.9	63.6	129.9	62.1	54.8	50.4	33.0	38.9	40.6	35.2	45.2	35.3	45.6	1.29
F	mg/L	0.1	0.1	0.1	0.1	0.0	0.1	0.1	0.0	0.1	0.7	0.1	0.1	0.1	0.1	1.03
Cl	mg/L	3.8	9.9	2.0	6.1	9.8	1.5	1.1	2.4	3.2	14.1	8.7	19.5	3.3	6.1	**1.86**
NO2	mg/L	0.9	0.5	0.7	0.3		0.5	0.2	1.8	0.5	1.0	1.5	0.7	0.5	0.7	1.27
Br	mg/L	0.4	0.2	0.2	0.1		0.2	0.2	0.4	0.2	0.2	0.0	0.3	0.2	0.2	1.07
NO3	mg/L	6.2	0.7	1.4	0.6	2.1	2.2	2.6	3.8	0.9	1.5	0.9	2.2	2.0	1.6	0.79
SO4	mg/L	10.6	11.0	42.1	251.5	11.7	16.0	6.8	78.4	19.0	251.8	58.0	57.0	12.0	59.8	**5.00**
HCO3	mg/L	191.0	163.5	232.5	353.9	231.9	193.4	175.1	255.1	153.2	275.2	379.8	233.1	184.9	255.1	1.38
Cr	µg/L	0.3	0.4	0.6	0.4	0.2	0.4	0.4	0.8	0.3	0.6	0.7	0.5	0.3	0.5	1.40
Mn	µg/L	0.1	1.1	0.6	0.2	0.3	0.6	0.1	0.1	0.2	0.3	1.5	3.9	0.2	0.3	1.46
Co	µg/L	0.0	0.0	0.1	0.7		0.0	0.1	0.0	0.1	4.4	0.7	0.5	0.0	0.4	**7.53**
Ni	µg/L	0.4	0.8	3.1	41.0	0.3	0.7	1.2	1.4	0.6	8.7	3.2	2.1	0.7	3.2	**4.82**
Cu	µg/L	0.0	2.3	17.2	11.4	0.3	0.4	11.7	7.0	1.0	34.8	16.6	6.5	0.5	11.8	**22.11**
Zn	µg/L	0.0	0.2	0.6	4.3	13.1	0.0	0.5	0.1	0.0	4.0	5.6	7.1	0.0	3.6	**120.34**
Rb	µg/L	0.2	0.2	0.3	1.3		0.3	0.2	0.2	0.3	9.6	2.6	1.9	0.2	1.3	**5.66**
Sr	µg/L	101.7	41.8	195.3	570.6	92.0	85.0	56.5	57.0	46.2	197.9	80.7	104.5	79.3	107.9	1.36
Ba	µg/L	9.9	74.4	20.6	16.8	33.0	11.2	4.4	4.4	10.2	16.4	34.2	38.9	10.8	19.5	**1.81**

Table 1: Sampling stations for mine water and reference water in the three study areas. Median values of physicochemical parameters based on six sampling campaigns between January and July 2024. Green columns represent reference stations; red columns represent mine water discharge stations (based on their location in the field). In addition, median values of all reference and mine waters were calculated to show the current influence of mines on water quality. Parameters whose concentration in the mine water is more than 50% higher than in the reference water are marked in bold.

sample name	sample date	δ34S [‰]	δ18O [‰]
M-MW1	4/11/2024	13,7	-11,6
M-MW2	4/11/2024	8,8	-5,7
S-MW2	4/11/2024	-6,6	-6,1
H-MW1	4/11/2024	-1,6	-8,5
H-MW2	4/11/2024	-2,6	-7,9
H-MW3	4/11/2024	-1	-9,8

Table 2: Sulphate isotope data of six mine waters, samples taken in April 2024. The stable isotope ratios represent the mean value of the repeat analyses of a sample (n ≥ 2). The values were accepted if the standard deviation of the repeat analysis was ≤ 0.8 for $\delta^{18}O$ and ≤ 1.0 for $\delta^{34}S$.

4.1. Data quality

As shown in Figure 3 72 % of the samples show an ion balance in the range of +/- 5 % and thus can be accepted as correct analyses. 19 % show ion balance errors in the range of +/- 5 % to +/- 10 %, while another 9 % of the samples show errors of more than +/- 10 %. As can be seen in Figure 3, there are both negative and positive outliers regarding electroneutrality. There is therefore no indication that essential ions were not analysed. For further processing and interpretations, all data sets with an error of more than +/- 10 % were removed from the dataset.

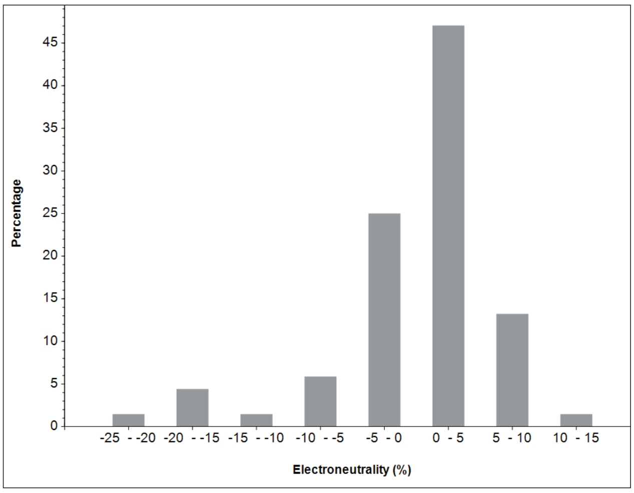

Figure 3: Ion balance of water samples. The electron neutrality of 72% of the samples lies within a range of +/- 5 %, a further 19 % lie between +/- 5 % and +/- 10 %, and in 9 % of the samples the error is greater than +/- 10 %. The analyses with an error >+/- 10 % were excluded from further processing.

4.2. Field parameters

The specific electrical conductivity (EC) (Fig. 4a) of the water ranges between 200 and 1050 µS/cm, with the reference samples (H-RW1, M-RW1, M-RW2, and S-RW1) always below a value of 350 µS/cm, while the mine waters (H-MW1, H-MW2, H-MW-3, M-MW1, M-MW2, S-MW2,) are significantly more mineralised with values above 400

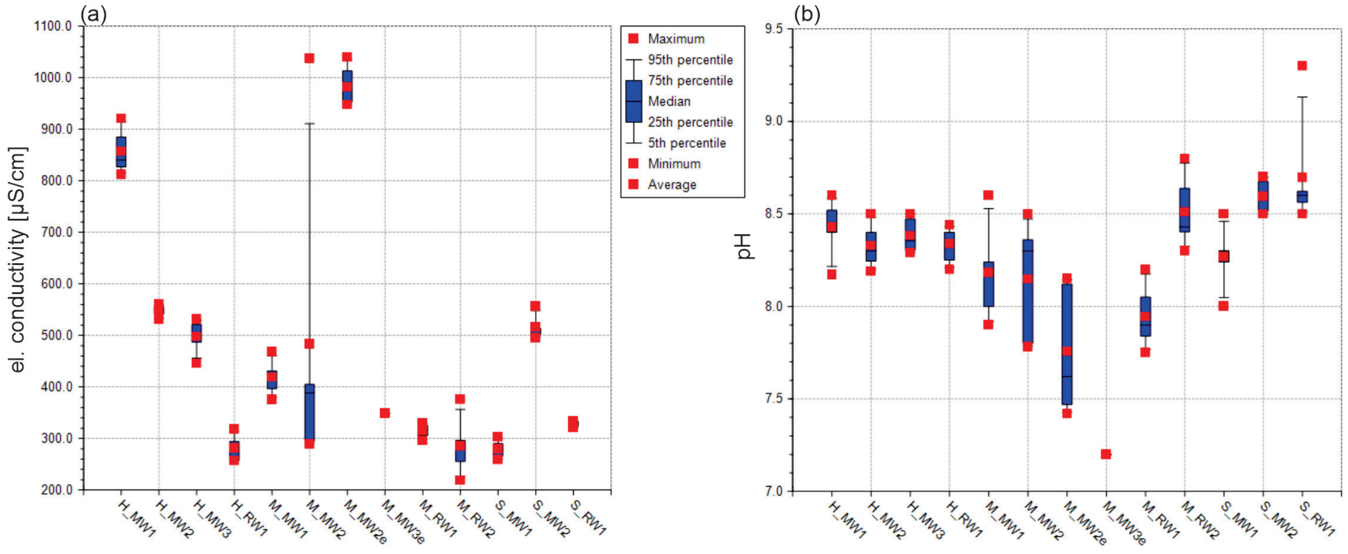

Figure 4: (a) Specific electrical conductivity and **(b)** pH values and their range over the observation period.

µS/cm. An exception is mine water S-MW1 which never exceeded 300 µS/cm. In M-MW2, significant fluctuations in EC between 300 and more than 1000 µS/cm were observed during the investigation period. The higher total mineralisations for this station are confirmed by external data (M-MW2e). Mine water H-MW1 shows high total mineralisation with a median value of 840 µS/cm and a minimum value of more than 800 µS/cm. Particularly noteworthy are the pH values (Fig. 4b), which are in the alkaline range, mostly between 8 and 8.5, both in the reference and in the mine water. During the study period, the values fluctuate at most stations but always remain well above the circumneutral range. The strongest fluctuations can also be seen in M-MW2. A single value well above a pH of 9 at monitoring site S-RW1 is considered a measurement error and thus as an outlier not considered for further interpretation. The external data (M-MW2e and M-MW3e) tend to show lower pH values, but also in the slightly alkaline range.

4.3. Major element distribution

The Piper plot in Figure 5 shows that the sampled waters are predominantly of the Ca-Mg-HCO_3 type and thus represent a typical groundwater composition in the Alpine region (Austrian Federal Environment Agency, 2024). This is true for all reference waters (green colors) and most of the mine waters (red, orange and purple colors) in each study area. The water composition at sample sites M-MW2 and H-MW1 differs clearly from that of the other mine waters. These two mine waters have comparably higher sulphate concentration, and additionally H-MW1 also has a higher sodium concentration. Consistent with the results of the field parameters (Fig. 4), M-MW2 has a fluctuating composition and can be classified as a Ca-HCO_3 or Ca-SO_4 water type, while H-MW1 can be classified as Na-SO_4 water type.

Interesting conclusions can be drawn from observing specific ion ratios (Fig. 6). The reference waters show the dominant dissolution processes in the natural aquifer, while ion ratios in the mine waters provide information about which further dissolution processes are anthropogenically induced. The waters can be classified according to their Ca/Mg ratios to obtain information about the types of carbonates or silicates available for dissolution in the different catchment areas. As shown in Figure 6a the Ca/Mg ratio of the reference waters indicates dolomite dissolution or calcium dissolution as dominating process. Mine waters in Mühlbach (circles) are calcium-dominated, while mine waters in Hüttau (rectangles) and S-MW2 in St. Veit (triangles) have an excess of magnesium, which can be explained with dissolution of magnesite as a relevant hydrochemical process.

The Na/Cl ratio (Fig. 6b) should be close to 1 if it is the result of dissolution of halite. As shown in Figure 6b, this is clearly the case for all reference stations and most mine waters. There are no significant differences in Na-Cl concentration between reference and mine waters. Only H-MW3 shows higher total salt concentration compared to all other stations. The values of M-MW2 and even more so those of H-MW1, on the other hand, show a noticeable sodium surplus.

The summed concentration of SO_4 and Cl versus HCO_3 can be used to distinguish between evaporite (or sulphide) dissolution and carbonate (or silicate) weathering as the main hydrochemical process. Looking at Figures 6c and d, it is obvious that M-MW2 and H-MW1 are again clearly different from all other stations. While most mine waters and all reference waters are dominated by HCO_3, these two stations show a clear excess of SO_4 and Cl, respectively. However, since both stations have a high Na/Cl ratio, it can be concluded that SO_4 is the dominant parameter, and thus either sulphate dissolution or sulphide oxidation are the driving forces causing the observed

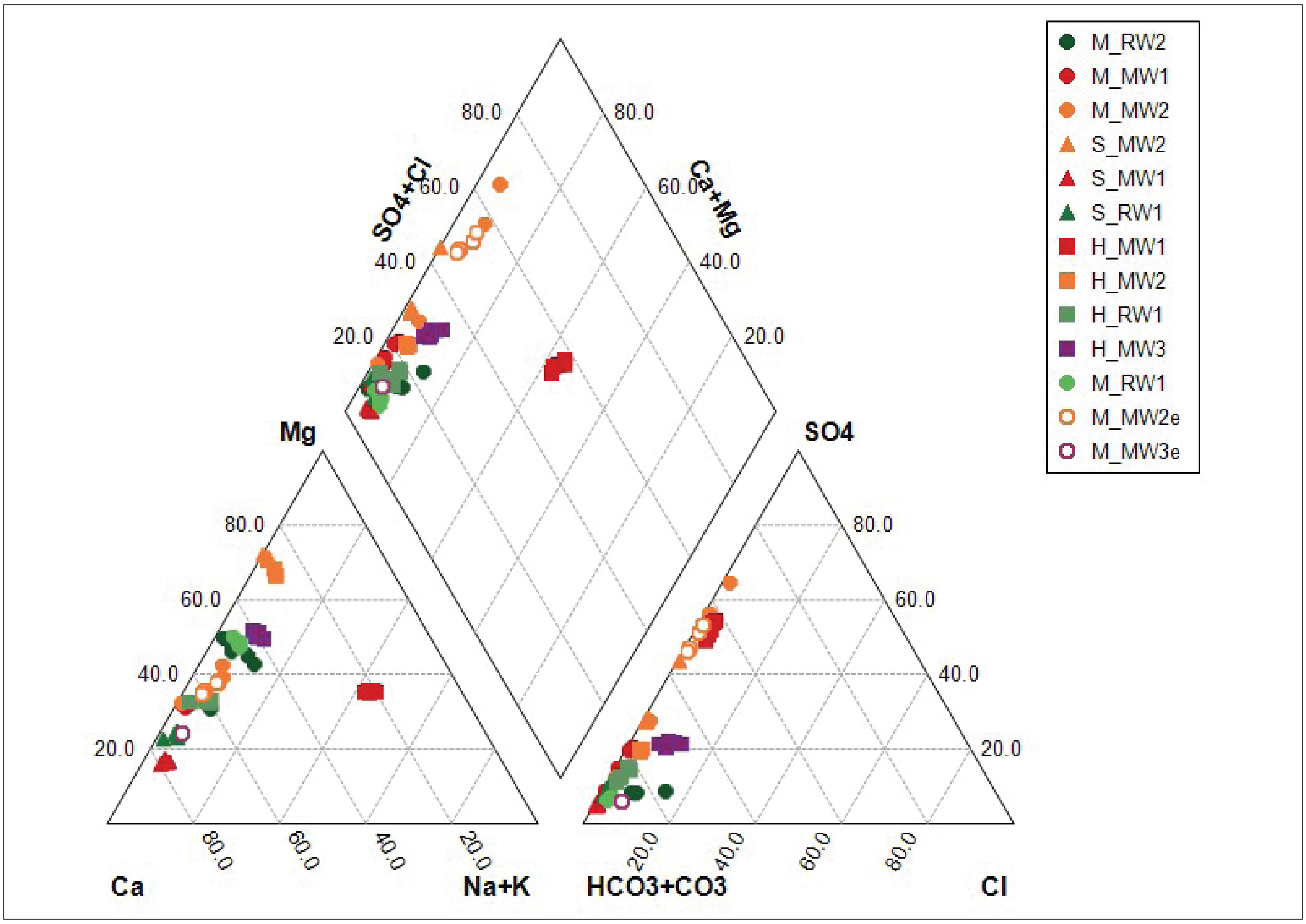

Figure 5: Major element composition of mine water (red colors) and reference water (green colors), symbols represent the three different mining regions: circles - Mühlbach, triangles - St. Veit, and rectangles - Hüttau. The open symbols represent data from external sources. The Piper plot shows the percentage distribution in meq/L and allows the waters to assign to specific water types according to Furtak und Langguth (1967).

water composition. The strong correlation of SO_4 with Ca + Mg (Fig. 6d) indicates that these ions originate from the same source, and the low ratio of 0.3 is evidence that generally sulphate dissolution is only a subordinate process in the natural catchments. Thus, major parts of the cations origin in carbonate or silicate solution. This is true for all stations except M-MW2 and H-MW1. Their higher sulphate fraction indicates an additional source of sulphate that is not available in the aquifer systems of the other stations and could probably be explained by the ongoing sulphide oxidation as described in formulae 1 and 2 in the copper mining systems.

4.4. Trace element concentration

The trace elements found at concentration above the detection limit are shown in Table 1 and Figure 7 as median values from five sampling campaigns (January to May 2024). Concentration of nickel (Ni), copper (Cu), zinc (Zn), strontium (Sr) and barium (Ba) were detected in the mine waters and can therefore be described as slightly enriched compared to the reference waters. The highest concentration of Ni is found in M-MW2, the highest concentration of Cu in H-MW1. Higher concentration of Sr is also found in both these sampling stations. However, all values are well below the World Health Organization's (WHO) limits for drinking water (World Health Organization, 2022).

4.5. Sulphate isotopes

The sulphate isotope signature shown in Figure 8 and Table 2 supports the assumption that the measured high sulphate concentration in mine waters is most likely due to sulphide oxidation. All mine water samples show negative $\delta^{18}O$ values between -11.6 ‰ (M-MW1) and -5,7 ‰ (M-MW2). This indicates that groundwater from the catchment area is incorporated into the sulphate molecules during sulphide oxidation. Although the values vary greatly, they are consistent with precipitation $\delta^{18}O$ isotope data for the region. The nearest ANIP (Austrian Network of Isotopes in Precipitation) station is located in Flachau (station ID IN50000046) (app. 50 km South of the study area) and provides values between -5 ‰ and -16 ‰ depending on the season (Austrian Federal Environment Agency, 2025). The wide range of variation reflects

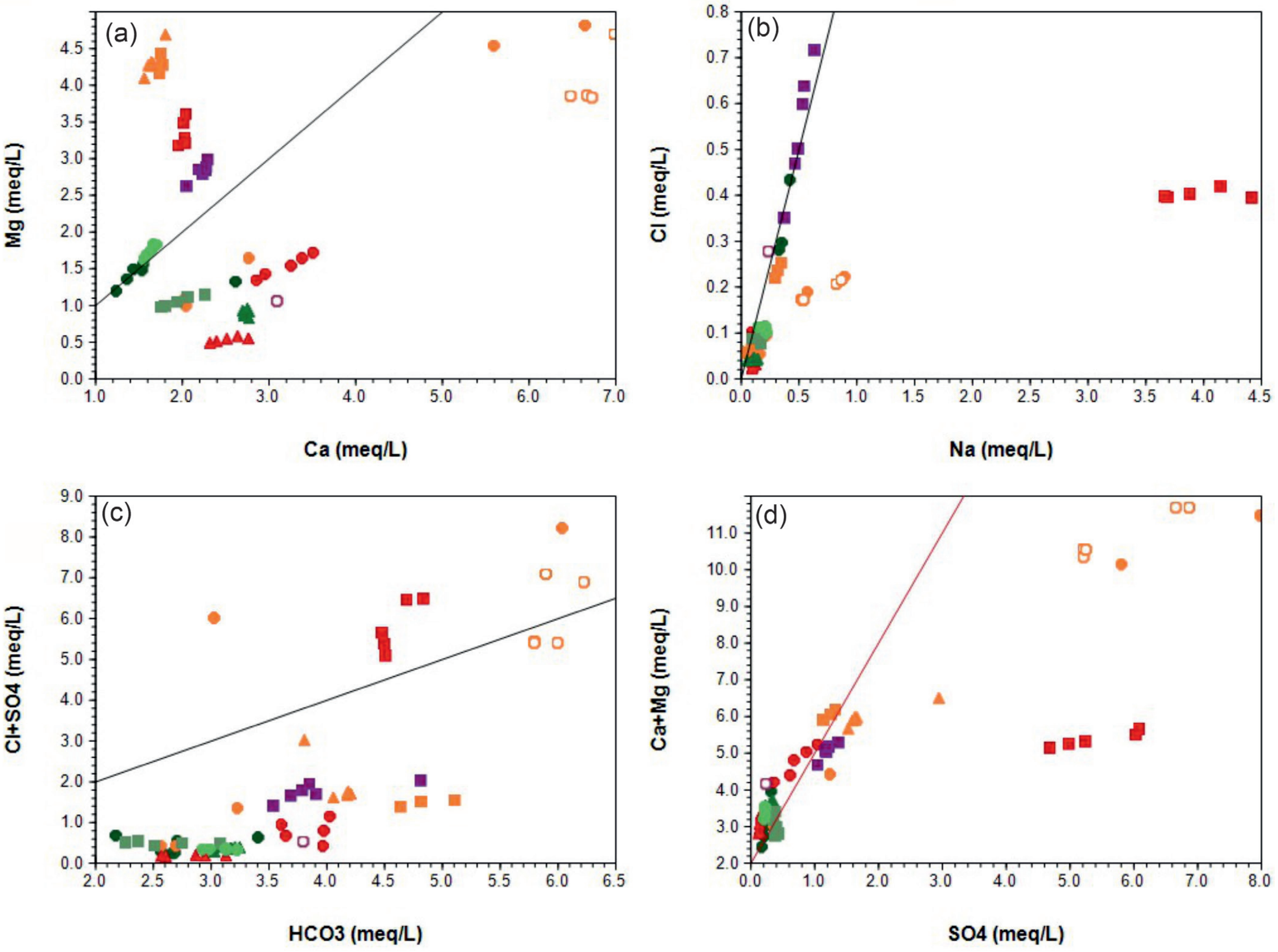

Figure 6: Scatter plots showing the correlation between **(a)** Ca/Mg, **(b)** Na/Cl, **(c)** HCO_3/SO_4+Cl and **(d)** SO_4/Ca+Mg. Black lines in (a), (b) and (c) represent the 1:1 lines, red line in (d) represents the linear trend line of the SO_4/Ca+Mg ratio of 0.3. See figure 5 for legend.

the differences in the average mean recharge altitude and the different hydrogeological conditions in the three catchment areas.

The δ^{34}S values are positive in Mühlbach, slightly negative in Hüttau and clearly negative in St. Veit and can be clearly attributed to the field of sulphide oxidation (Fig. 8). The differences reflect the varying ore compositions in the three mining areas.

5. Discussion

The investigation of seven mine water outflows and six unaffected reference waters in the catchment areas of the mining regions Mühlbach, Hüttau and St.Veit provides information on whether and to what extent anthropogenic interventions in the hydrogeological system of the abandoned mines can still be detected after decades without any mine water treatment. The comparison of the field parameters EC and pH shows first surprising results. The total mineralization of the stations identified as mine water is considerably, but not dramatically, higher than that of the reference stations. With S-MW1, a mine water inflow in the Sunnpau mine, St. Veit proves to be the least mineralized water of all.

- Sulphide weathering has been proven to occur at sampling stations in all three mines. However, no acidification is observed. There are two possible reasons for this: firstly, chalcopyrite is the main ore in the deposits examined. Although its weathering can lead to acidification of mine water, this occurs to a much lesser extent than would be expected with pyrite weathering. Secondly, the measured alkaline pH values can be explained by efficient buffering processes in the surrounding rock. At all three study sites typical ore veins are embedded in carbonates of the magnesite-siderite and dolomite-ankerite series (Feitzinger et al. 1998). Although none of the analyzed samples can be classified as acid mine drainage (AMD) according to Gray (1996) or Kuma et al. (2011) the hydrochemical data show clear differences between mine waters and reference waters. This applies both to the hydrochemical signature and to their absolute concentration, which is generally higher in mine waters than in reference waters. Dissolution of toxic metals in mining environments is aided by an acid-

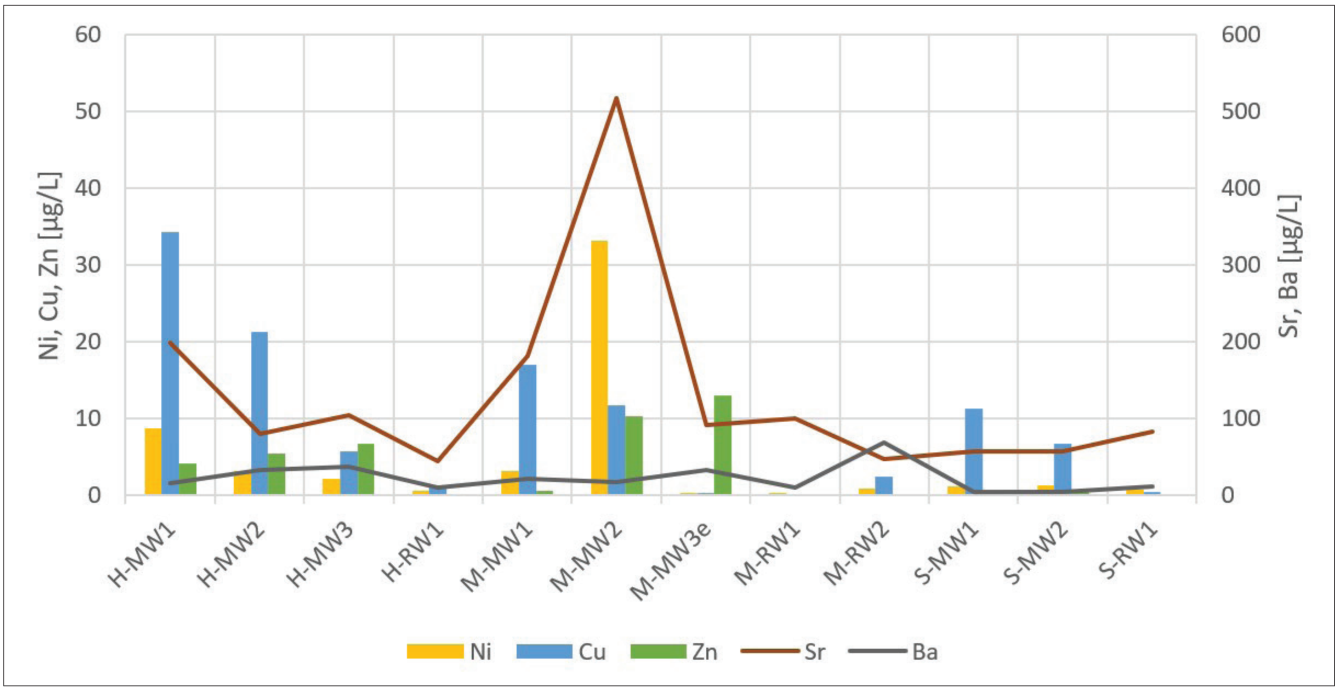

Figure 7: Mean values (n = 5) of the detected trace element concentration in reference and mine water stations. Columns represent Ni, Cu and Zn concentration, lines represent the one order higher concentration of Sr and Ba.

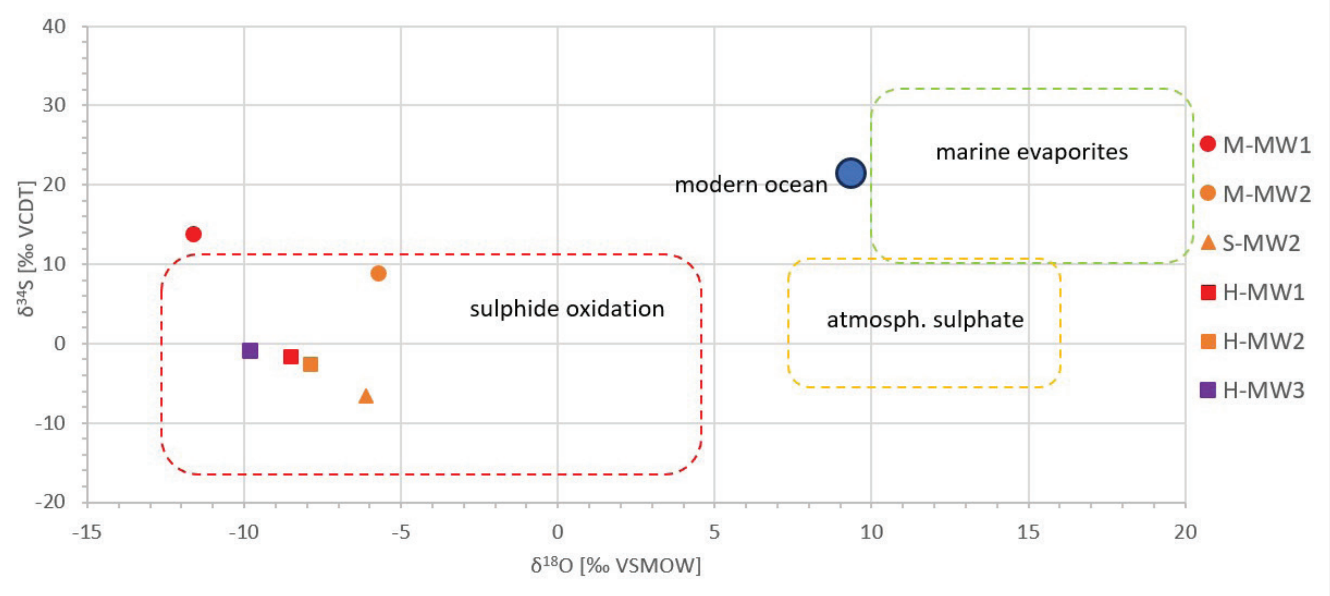

Figure 8: Isotopic signature of sulphate in mine waters, sampling campaign April 2024. All samples are isotopically light with respect to $\delta^{34}S$ and $\delta^{18}O$ and are plotted in a range that is typical for sulphates resulting from sulphide oxidation (Wang and Zhang, 2019).

ic environment (Nordstrom, 2011). Thus, the overall low concentration of heavy metals and metalloids is consistent with the observed alkaline pH values. In addition to these general statements on the comparison of mine water and reference water, the results of three distinct sampling sites are particularly striking:

- Station S-MW1 (St. Veit), is always in the range of reference waters and even has the lowest total mineralization of all stations. Only the trace element concentration is slightly elevated compared to the reference waters. S-MW1 is a spring that emerges from a fissure in the adit (Fig. 2f). It was sampled before it was affected by mining. It is therefore actually more of a perfect reference water than a mine water.

- Station M-MW2 (Mühlbach) is characterized by large fluctuations in its hydrochemical composition. While some samples exhibit typical Ca-Mg-HCO_3 waters, others show Ca-Mg-SO_4 water types. The fluctuations manifest themselves in the sulphate content as well as in the total mineralization. The major element composition provides strong evidence for the origin of the elevated sulphate concentration in the sulphides of the ore deposits, which have been exposed to increased weathering due to mining activities. This interpretation is supported by the results of sulphate isotope analysis. Generally light isotope signatures can be easily reconciled with the process of sulphide weathering. The discharge at the sampling site was not accurately measured during sampling. As can be seen in Figure 2a, this would have required structural measures at the tunnel entrance that would have exceeded the time and financial scope of this study. However, semiquantitative discharge estimates during sampling indicate that high sulphate concentration is associated with high discharge and low mineralized water with low flow conditions. Normally, one would expect a dilution effect at higher discharge values due to the short-term influence of low mineralized rain water. In M-MW2 the opposite seems to be the case. This can be interpreted as follows: The adits are hydrogeologically similar to a karst system (Bedoya-Gonzalez et al., 2021). During high flows, the water level rises and floods areas that are not flooded during low flows. In these transiently flooded adits, the process of sulphide oxidation is not complete, and leaching can still occur, while these hydrochemical processes are largely completed in the permanently flooded adits.

- In station H-MW1 (Hüttau) a very special hydrochemical composition was found. Permanently high total mineralization is associated with high sulphate and high sodium concentration. The sulphate concentration is not matched by adequate calcium content, and the sodium is not balanced by chloride. The sulphate concentration can be explained by sulphide oxidation as discussed for M-MW2. To explain the origin of sodium, a linear regression analysis with sodium as the dependent variable was performed and is shown in Supplement 2. It shows a significant positive correlation between sodium and magnesium, sulphate and bicarbonate and no correlation with chloride and potassium in the waters of station H-MW1. In contrast, the reference waters and all other mine waters show a clear correlation with chloride and potassium. It can be concluded that hydrochemical processes occur in the Hüttau catchment that do not occur in any of the other investigated areas, despite comparable geological conditions. Since there is only a non-significant negative correlation with potassium, cation exchange can be largely excluded as an explanation. Silicon has not been analysed and therefore thermodynamic models cannot calculate silica mineral phases. However, weathering of silicate minerals could be a possible explanation.

6. Conclusion

The presented study provides an overview of the existing mine water outlets and the remaining effects of mining on the quality of springs and watercourses in the Ore of the Alps UGG region. It adds a new aspect to the topic of historical mining in the Geopark. An important result of the study for the Geopark region is the generally low contamination of mine waters with pollutants. A previously unnoticed environmental hazard could thus be ruled out.

However, it is evident that even decades after mine closure, a clear influence of mining can still be seen in the mine waters. While leaching is largely complete, sulphide weathering in the artificially perforated rock is still a relevant process. Sulphate is the clearest indicator parameter for this.

The Greywacke Zone, the geological framework of the mines, is mainly characterised by silicate metamorphic rocks without considerable buffer capacity. However, the mineralization itself is associated with carbonate rocks, in particular magnesite-siderite and dolomite-ankerite carbonates (Feitzinger et al., 1998). Dissolution of these ore-associated carbonates keeps the pH in the alkaline range despite sulphide oxidation, thus preventing the release of toxic metals via the groundwater pathway. Although not yet verifiable with exact discharge measurements, field observations indicate that during periods of high flow, Ni concentration is elevated in M-MW2. In particular, the fluctuation of the concentration in M-MW2 with discharge indicates that major leaching and oxidation processes have been completed in most of the permanently flooded mine water outlets, while it continues in only temporarily flooded parts of the mine.

As with all research, the results of this project raised new questions. Explanations for the very specific hydrochemistry found at sampling site H-MW1 remain open. Detailed geological-hydrogeological, hydrochemical and geochemical investigations in the catchment area of the Larzenbach copper mine in Hüttau are required for this purpose.

Further regular sampling, including continuous discharge recordings especially at M-MW2 in combination with detailed geological-hydrogeological mapping of the catchment area could contribute significantly to a better understanding of the mine water dynamics in Mühlbach.

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