

Review

The Lengenbach Quarry in Switzerland: Classic Locality for Rare Thallium Sulfosalts [†]

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Abstract: The Lengenbach quarry is a world-famous mineral locality, especially known for its rare and well-crystallized Tl, Pb, Ag, and Cu bearing sulfosalts. As of June 2018, it is the type locality for 44 different mineral species, making it one of the most prolific localities worldwide. A total of 33 thallium mineral species have been identified, 23 of which are type minerals. A brief description of several thallium species of special interest follows a concise and general overview of the thallium mineralization.

Keywords: Lengenbach; Binn valley; thallium; sulfosalts; hutchinsonite; fangite; richardsollyite; sartorite; routhierite-stalderite; chabournéite-dalnegroite

1. Introduction

The Lengenbach quarry in the Binn valley, Valais, Switzerland (Figures 1 and 2) is located in Triassic meta-dolomites of the Penninic zone in the Swiss Alps. Metal extraction for economic purposes never occurred in the quarry, but specimen extraction has been continuously carried out since 1958. The quarry is currently operated by the Forschungsgemeinschaft Lengenbach (FGL, literally: Lengenbach Research Association), financed by a group of idealistic collectors and by the local community of Binn. The purpose of the research association is to promote scientific research on the unique minerals of the Lengenbach deposit and of other dolomite localities in the Binn valley. An intermittent, measured specimen extraction during the snow-free summer months shall guarantee the potential for scientific investigations on the one hand and deliver dolomite material for a publicly accessible dump, serving as an attraction to equally eager tourists and mineral collectors, on the other hand.

This brief review gives a glimpse at the current status of the mineralogical research with regard to the thallium mineralization at the locality. For further information about history, geology, and mineral extracting work we recommend References [1,2].



Figure 1. The upper part of the Lengenbach quarry in the Binn valley, view to the east.



Figure 2. Ralph Cannon, technical head of the Forschungsgemeinschaft Lengenbach (FGL) research association, at the entrance of the quarry in front of the concrete hall, where the current mineral-extraction activities are carried out at the lowest dolomite level.

2. Geochemical Setting and Formation of the Lengenbach Locality

The Lengenbach ore body is located within the Penninic Monte Leone nappe, at the Northern front and subvertical hinge zone of a large fold. The stratabound mineralization occurs in the stratigraphically uppermost part of the 240 m thick dolomite sequence.

The formation of the highly complex mineralization in the Lengenbach deposit is not yet completely understood. While Graeser [3] suggested a late introduction of As, Tl, and Cu into a pre-existing Fe-Pb-Zn mineralization during Alpine metamorphism from the underlying gneissic

basement, Hofmann and Knill [4] proposed a pre-Alpine origin of those elements and a subsequent isochemical Alpine metamorphism, under upper greenschist to lower amphibolite facies.

According to Hofmann and Knill [4], the distinct mineral associations in the different parts of the Lengenbach dolomite can be understood as a result of slow crystallization processes in two different redox environments. One is based on graphite and/or pyrite–pyrrhotite, leading to zerovalent arsenic. The other, which was essential for the rare sulfosalts' formation, is controlled by baryte (sulfate)–pyrite (sulfide), leading to trivalent arsenic. Accordingly, the As(III)-rich zone in the central part of the quarry shows an enrichment in baryte and hosts the coveted Tl–Pb–Ag–Cu bearing sulfosalts.

Graeser [3] as well as Hofmann and Knill [4] have each proposed a zonation scheme for the different types of mineral assemblages. While the former considers in essence only mineralogical and spatial criteria, the later rely on geochemistry and there is no obvious link between the two zonations. However, it is clear that the Tl-rich zone is restricted to the central part of the quarry. While on a broad scale the different bedding-parallel zones containing the different assemblages strike subvertically in an east–west direction (Figure 1), on the more local scale, they can be subdivided into ribbons and ellipsoidal lenses that thicken, to a maximum thickness of 0.5 m, and pinch out.

The FGL has been working for a few years on three such ribbons in the Tl-rich central zone (Figure 3). They are spaced approximately 1 m apart, measure a maximum of 4 m × 2 m and are designated, from north to south as ribbons 1, 1/2, and 2. Structurally, they are ellipsoidal in shape, with a sharp contact to the surrounding, mineral-poor dolomite. The contrast is essentially a mineralogical-geochemical, not a lithological contrast. Thanks to their high realgar contents, all three ribbons are easily identified in situ. But while ribbon 1 is very rich in thallium species, the very brittle and orpiment-rich dolomite of ribbon 1/2 is poorer in Tl while the realgar-richest ribbon 2 is almost bare of any thallium-species. Table 1 summarizes all species found in ribbon 1; 18 species containing thallium as one of main constituents are marked in bold characters.



Figure 3. Three realgar-rich ribbons in the central dolomite zone in the lower part of the quarry. Ribbon 1 (red) is very rich in thallium species, ribbon 1/2 (yellow) contains much orpiment in a brittle dolomite and is poorer in thallium, ribbon 2 (orange) is the realgar-richest ribbon, but almost bare of any thallium species. The maximum ribbon thickness is about 0.5 m. Dashed lines show the expected extension of the ribbons below the debris.

Table 1. Species constituting the mineral assemblage of ribbon 1 in the Lengenbach quarry. Tl minerals are marked in bold, modified after reference [5].

“Adularia” and “hyalophane”	Hutchinsonite	Rathite
Aktashite	Hydrocerussite	Realgar
Argentobaumhauerite	Incomsartorite	Richardsollyite
Argentodufrenoyseite	Imhofite	Routhierite
Argentoliveingite	Jentschite	Rutile
Arsenic	Jordanite	Sartorite
Baryte	Kaolinite	Seligmannite
Baumhauerite	Lengenbachite	Silver
Bernardite	Liveingite	Sinnerite
Canfieldite (Te-rich)	Marrite	Smithite
Cerussite	Muscovite	Sphalerite
Coffinite	Nowackiite	Stalderite
Covellite	Orpiment	Tennantite
Dolomite	Pararealgar	Thalcusite
Dravite	Parapierrotite	Thorite
Dufrenoyseite	Picotpaulite	Tochilinite
Edenharterite	Proustite	Trechmannite
Erniggliite	Pyrite	Uraninite
Ferrostalderite	Quadratite	Wallisite
Galena	Quartz	Wurtzite
Hatchite	Ralphcannonite	Xanthoconite

3. Overview of Thallium Minerals at Lengenbach

As of June 2018, the Lengenbach quarry hosts 160 different mineral species, with sulfides and sulfosalts being the major group representing 57% of these species (Table 2 and Figure 4). Forty-four minerals are so called type minerals as they have been found and described for the first time from this locality. Twenty-three of them are thallium minerals. Table 3 lists all known Lengenbach thallium minerals as of today.

Table 2. Mineralogical overview of the Lengenbach quarry.

Category	Number
Mineral species *	160
Elements	6
Sulfides and sulfosalts	91
Chalcogenides	2
Carbonates	10
Sulfates and molybdates	10
Phosphates and arsenates	8
Oxides	13
Silicates	20
Type minerals	44
Thallium minerals	33

* IMA (International Mineralogical Association) approved.

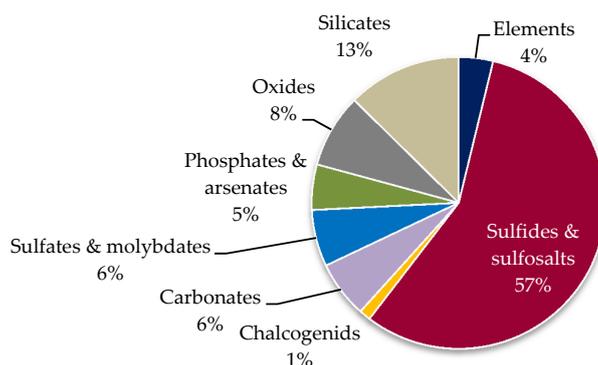


Figure 4. Distribution of mineral classes at Lengenbach.

Table 3. Thallium minerals at Lengenbach.

Mineral Species	Chemical Formula	Year of First Description from Lengenbach	Reference for the Occurrence at Lengenbach
Bernardite	$Tl(As,Sb)_5S_8$	1992	Graeser (in Hofmann et al.) [6]
Chabournéite	$Tl_4Pb_2(Sb,As)_{20}S_{34}$	2017	Roth [7]
Dalnegröite *	$Tl_4Pb_2(As,Sb)_{20}S_{34}$	2009	Nestola et al. [8]
Dekatriasartorite *	$TlPb_{58}As_{97}S_{204}$	2017	Topa et al. [9]
Edenharterite *	$TlPbAs_3S_6$	1992	Graeser & Schwander [10]
Enneasartorite *	$Tl_6Pb_{32}As_{70}S_{140}$	2017	Topa et al. [11]
Ernigglite *	$Tl_2SnAs_2S_6$	1992	Graeser et al. [12]
Fangite	Tl_3AsS_4	2017	Roth [13]
Ferrostalderite *	$CuFe_2TlAs_2S_6$	2016	Biagioni et al. [14]
Gabrielite *	$Tl_2AgCu_2As_3S_7$	2006	Graeser et al. [15]
Hatchite *	$AgTlPbAs_2S_5$	1912	Solly & Smith [16]
Hendekasartorite *	$Tl_2Pb_{48}As_{82}S_{172}$	2017	Topa et al. [11]
Heptasartorite *	$Tl_7Pb_{22}As_{55}S_{108}$	2017	Topa et al. [11]
Hutchinsonite *	$TlPbAs_5S_9$	1905	Solly [17]
Imhofite *	$Tl_{5.6}As_{15}S_{25.3}$	1965	Burri et al. [18]
Incomsartorite *	$Tl_6Pb_{144}As_{246}S_{516}$	2016	Topa et al. [19]
Jentschite *	$TlPbAs_2SbS_6$	1997	Graeser & Edenharter [20]
Lorándite	$TlAsS_2$	1967	Graeser [21]
Parapierrotite	$Tl(Sb,As)_5S_8$	2016	Raber [22]
Philrothite *	$TlAs_3S_5$	2014	Bindi et al. [23]
Picotpaulite	$TlFe_2S_3$	2015	Roth [24]
Raberite *	$Tl_5Ag_4As_6SbS_{15}$	2012	Bindi et al. [25]
Raguinite	$TlFeS_2$	2017	Roth [26]
Ralphcannonite *	$AgZn_2TlAs_2S_6$	2015	Bindi et al. [27]
Rathite *	$Ag_2Pb_{12-x}Tl_{x/2}As_{18+x/2}S_{40}$	1896	Baumhauer [28]
Richardsollyite *	$TlPbAsS_3$	2017	Meisser et al. [5]
Routhierite	$CuHg_2TlAs_2S_6$	2016	Roth [29]
Sicherite *	$TlAg_2(As,Sb)_3S_6$	2001	Graeser et al. [30]
Spaltiite *	$Tl_2Cu_2As_2S_5$	2014	Graeser et al. [31]
Stalderite *	$CuZn_2TlAs_2S_6$	1995	Graeser et al. [32]
Thalculusite	$Tl_2Cu_3FeS_4$	2005	Cannon [33]
Wallisite *	$CuTlPbAs_2S_5$	1965	Nowacki [34]
Weissbergite	$TlSbS_2$	2014	Raber & Roth [35]

* Type minerals from the Lengenbach quarry.

From the 73 valid mineral species containing essential thallium worldwide (according to mindat.org [36]), the striking number of 33 species (45.2%) could be found at Lengenbach.

4. On Some Special Thallium Minerals from the Lengenbach Quarry

We focus here on a few thallium species of special interest, as a discussion of all thallium sulfosalts would be beyond the scope of this brief note.

4.1. Hutchinsonite—The First of Its Kind

The first thallium mineral from Lengenbach, hutchinsonite, $TlPbAs_5S_9$, was found in 1903, when the English expert of Lengenbach minerals, Richard Harrison Solly (1851–1925), during one of his many trips to this remote Binn valley, recognized that the red to greyish-black, often flattened orthorhombic crystals probably were belonging to a new species. He briefly described it in 1904 [37], without giving it a name. In 1905, his colleague at the British Museum, G.T. Prior was able to reveal the presence of 20 wt % Tl in hutchinsonite. This was of “especial interest”, as Solly [17] wrote in his following detailed description, in which he named the mineral after Arthur Hutchinson (1866–1937). Prior’s discovery of thallium in hutchinsonite was important enough to result in a short note in *Nature* [38], as it was only the third mineral worldwide (after crookesite and lorándite) to contain thallium as an essential constituent.

Hutchinsonite is the most common thallium species in the deposit and represents the type structure of a family of complex sulfosalts. Its crystals are commonly transparent and prismatic, due to an elongation parallel to the *c* axis (see Figures 5–7), or, rarely, more or less isometric. Hutchinsonite may contain antimony that contributes the crystals to be darker and opaque. The two varieties, prismatic Sb-free and more isometric Sb-bearing hutchinsonites, may be closely associated. The crystals reach 2 mm in size.



Figure 5. Dark wine-red, transparent, lath-like hutchinsonite crystals on dolomite. Field of view 1.4 mm. Photo: Edgar Müller.



Figure 6. Translucent aggregate of parallel-oriented, red, flattened hutchinsonite crystals with pyrite on dolomite. Field of view: 2.5 mm. Photo: Stephan Wolfsried.



Figure 7. Prismatic dark-red to grey crystals of hutchinsonite. Field of view: 1.5 mm. Photo: Stephan Wolfsried.

4.2. Fangite—The Thallium-Richest

Well-developed crystals of fangite, Tl_3AsS_4 , could recently be identified for the first time [13]—to our knowledge, the first microscopically visible crystals of this species worldwide. They are small, deep red, and very shiny (Figure 8). With more than 75 wt % Tl, fangite is the thallium-richest mineral discovered at Lengenbach up to now.

A morphological study of the small crystals [26] showed them to display different combinations of crystal forms, irrespective of the fact that the prismatic habit is always approximately the same (Figure 9).



Figure 8. Tiny, prismatic, lustrous, red fangite crystals on dolomite. Field of view: 0.75 mm. Photo: Mischa Crumbach.

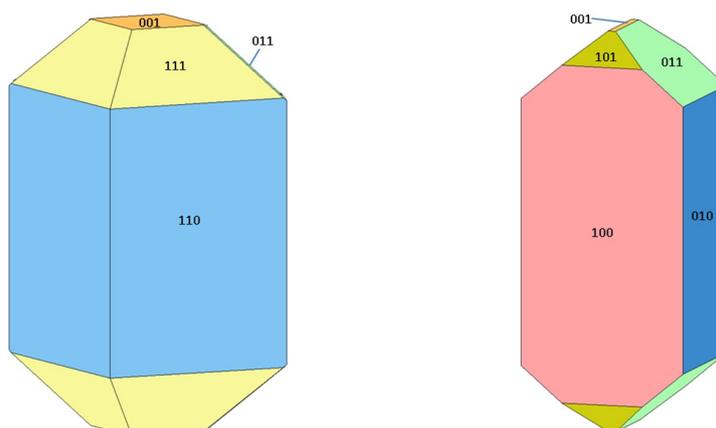


Figure 9. Fangite crystals showing different combinations of forms but similar habits. A distinct color is assigned to each crystal form. The Miller indices of the different forms are given without brackets FACES drawings [39].

4.3. Richardsollyite—Honoring a Pioneer

In 2015, the FGL extracted two specimens with an unknown mineral from the very Tl-rich dolomite ribbon 1 (Figure 3) in the center of the quarry. Its chemistry, as first determined by Energy Dispersive X-ray Spectroscopy (EDXS) measurements in two independent institutes, showed the presence of Tl, Pb, As, and S in a simple, yet unknown ratio of 1:1:1:3. Also the powder X-ray diagram did not match with any known listed natural or synthetic chemical compound in the relevant databases. One year later, Meisser et al. [5] could describe this mineral as a new species with the name richardsollyite (Figure 10), TlPbAsS_3 , honoring the aforementioned pioneer of Lengenbach investigations at the dawn of the twentieth century, R.H. Solly. The crystal structure of richardsollyite is new in nature, being previously known only in some synthetic alkali sulfosalts [5].



Figure 10. Holotype specimen of richardsollyite (dark grey) with hutchinsonite (dark red) and realgar. Field of view: 3.5 mm. Mineralogical Collection of the Musée cantonal de géologie (MGL no. 080126), photo: Stefan Ansermet.

4.4. The New “Sartorites”—From a Species to a Group

Sartorite, PbAs_2S_4 , was first described by vom Rath as “scleroclase” in 1864 [40], and shortly after renamed by Dana [41] to honor Wolfgang Sartorius von Waltershausen (1809–1876), a professor of mineralogy in Göttingen, Germany. In 1919, Smith and Solly [42] recognized that sartorite has—despite its simple chemical formula—a quite unique and complex crystal structure: “sartorite appears to rank with the telluride of gold, calaverite, in the peculiarity of its atomic arrangement, since in certain at least of his crystals there exist simultaneously two or even three incongruent space-lattices, which may be supposed derivable from one another by a slight shear.” It is quite remarkable that they were able, with the limited methods of their time—in essence only crystal-morphological investigations—to recognize the so-called complex and incommensurate nature of both the calaverite and, partly, sartorite structures.

After the introduction of X-ray investigations in crystallography, several different monoclinic super cells (superstructures) were described [43–46]. Berlepsch et al. [46] pointed to the crystallographic consequences of the complex correlated atomic substitution by which substantial amounts of thallium are incorporated into the sartorite structure. They found a so-called 9-fold superstructure for a sartorite with up to 6.5 wt % Tl and discussed this as a “lock-in” structure with a commensurate lattice, for a species that they regarded as usually incommensurate.

The true nature of sartorite was finally resolved by Topa et al. [9,11,19]. Based on a systematic combination of electron microprobe measurements and crystal structure determinations they showed that “sartorite” actually represents a group of different mineral species with distinct crystal structures and distinct chemical compositions. According to their different superstructures, the new “sartorites” were named by adding a Greek prefix, which corresponds to an integral multiple of the basic “sartorite” substructure with 4.2 Å (Table 4).

Table 4. The new sartorite minerals.

Mineral Name	Chemical Formula	Superstructure of the Sartorite Subcell (4.2 Å)
Heptasartorite [11]	Tl ₇ Pb ₂₂ As ₅₅ S ₁₀₈	7-fold
Enneasartorite [11]	Tl ₆ Pb ₃₂ As ₇₀ S ₁₄₀	9-fold
Hendekasartorite [11]	Tl ₂ Pb ₄₈ As ₈₂ S ₁₇₂	11-fold
Incomsartorite [19]	Tl ₆ Pb ₁₄₄ As ₂₄₆ S ₅₁₆	11-fold (incommensurate)
Dekatriasartorite [9]	TlPb ₅₈ As ₉₇ S ₂₀₄	13-fold

These species are not visually distinguishable. The crystals are lead-grey with a metallic luster. Well-developed crystals show a prismatic habit with more or less distinct striation parallel to the longitudinal axis (Figure 11). Their length reaches several centimeters.

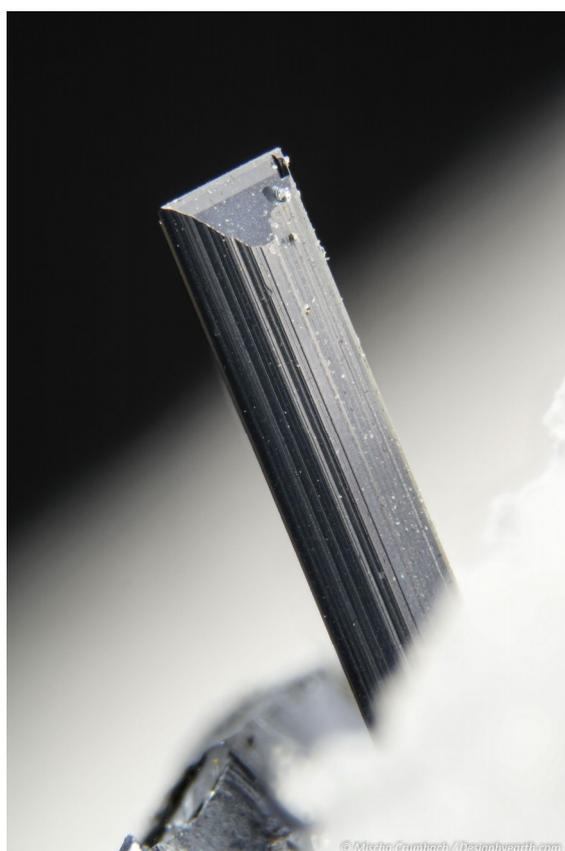


Figure 11. Lead-grey, metallic, prismatic crystal of dekatrisartorite. Field of view 4.8 mm. Photo: Mischa Crumbach.

4.5. The Routhierite-Stalderite Group—Complex Substitutions

The routhierite-stalderite series is a group of thallium arsenio-sulfosalts with, in addition, monovalent (Me1: Cu⁺, Ag⁺) and bivalent metal ions (Me2: Hg²⁺, Zn²⁺, Fe²⁺) and the generic formula TlMe1⁺Me2²⁺As₂S₆. Accordingly, six different combinations, and thus six distinct mineral species are theoretically possible in this group (Table 5, Figure 12). Five could indeed be found in nature: arsiccioite, routhierite, stalderite, ralphcannonite, and ferrostalderite. The latter four occur in the Lengenbach quarry, the type-locality for the latter three. Routhierite and arsiccioite are red to dark-red in color, the other three are dark grey with a metallic luster, but they all show the same pseudo-cubic to prismatic morphology (≤1 mm).

Table 5. Routhierite-stalderite group minerals.

Mineral Species	Chemical Formula
Routhierite	TlCuHg ₂ As ₂ S ₆
Stalderite	TlCuZn ₂ As ₂ S ₆
Ferrostalderite	TlCuFe ₂ As ₂ S ₆
Ralphcannonite	TlAgZn ₂ As ₂ S ₆
Arsiccioite	TlAgHg ₂ As ₂ S ₆
Unknown	TlAgFe ₂ As ₂ S ₆

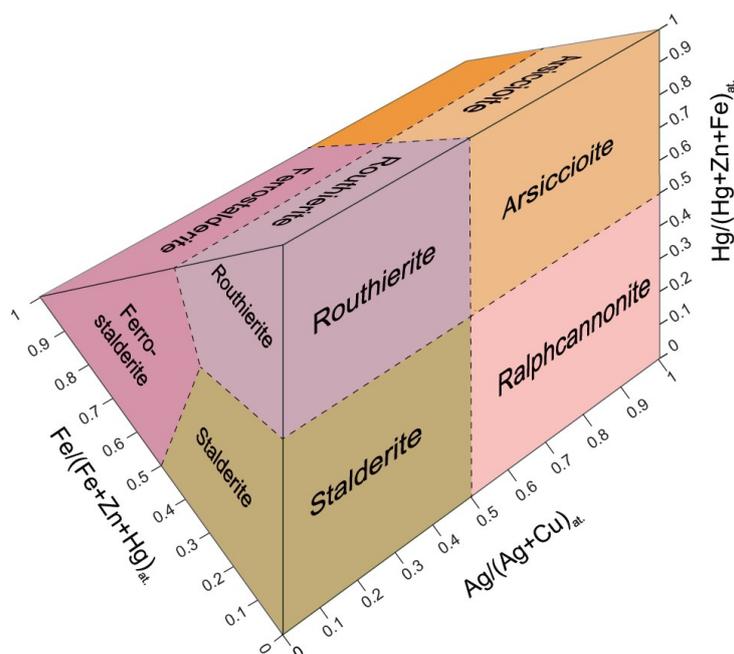


Figure 12. The minerals of the routhierite-stalderite group in a block diagram. The six distinct species are the result of the six possible combinations of monovalent (Ag^+ , Cu^+) and bivalent metal ions (Zn^{2+} , Hg^{2+} , Fe^{2+}). Modified after Reference [27].

In the frame of a series of EDXS analyses on Lengenbach samples from the mineralogical collection of the Eidgenössische Technische Hochschule (ETH) collection in Zurich, we recently identified what to our knowledge are the first completely idiomorphic crystals of routhierite worldwide (Figure 13).

Arsiccioite has not yet been found at Lengenbach. However, a single EDXS analysis of a minute crystal indicated a possible dominance of silver and iron, and thus, the possible existence of the last unreported member of this group in the quarry.

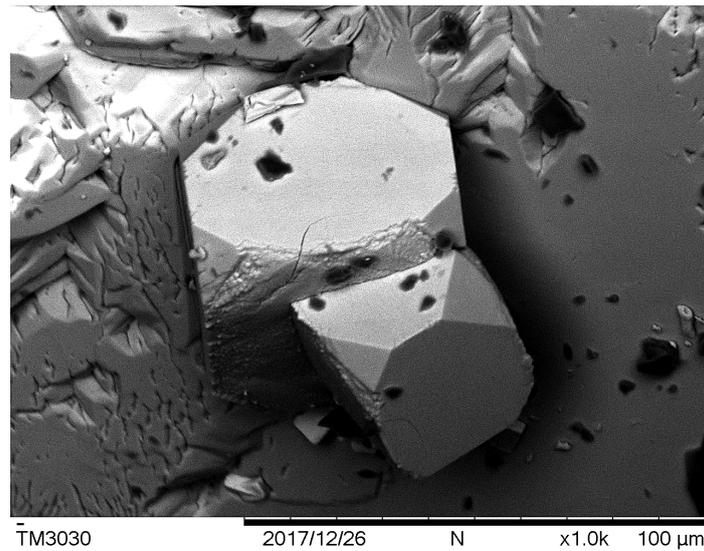


Figure 13. Idiomorphic, pseudo-cubic crystals of routhierite from the mineralogical collection of the Eidgenössische Technische Hochschule ETH in Zurich.

4.6. Chabournéite-Dalnegroite—And Possibly More

Dalnegroite was described as a new mineral species from Lengenbach in 2009 [8]. It is considered the As-analogue of chabournéite. The latter was also recently identified in one single specimen showing what appears to be the first distinct crystals for the species: They are very similar to the dalnegroite crystals and also show a strong similarity to lengenbachite crystals, which may imply that the two rare species may have been mistaken for lengenbachite in the past (Figure 14). Both dalnegroite and chabournéite are Pb-bearing species. In ribbon 1/2, however, the FGL has recently found a few samples of a Pb-free dalnegroite (Figure 15), the equivalent of the Pb-free chabournéite from Jas Roux, France [47,48]. The Pb-free species may well be distinct minerals (Figure 16). Their study needs to be refined and completed.



Figure 14. Black, stellar aggregates of chabournéite on dolomite. Field of view 1.2 mm. Photo: M. Crumbach.

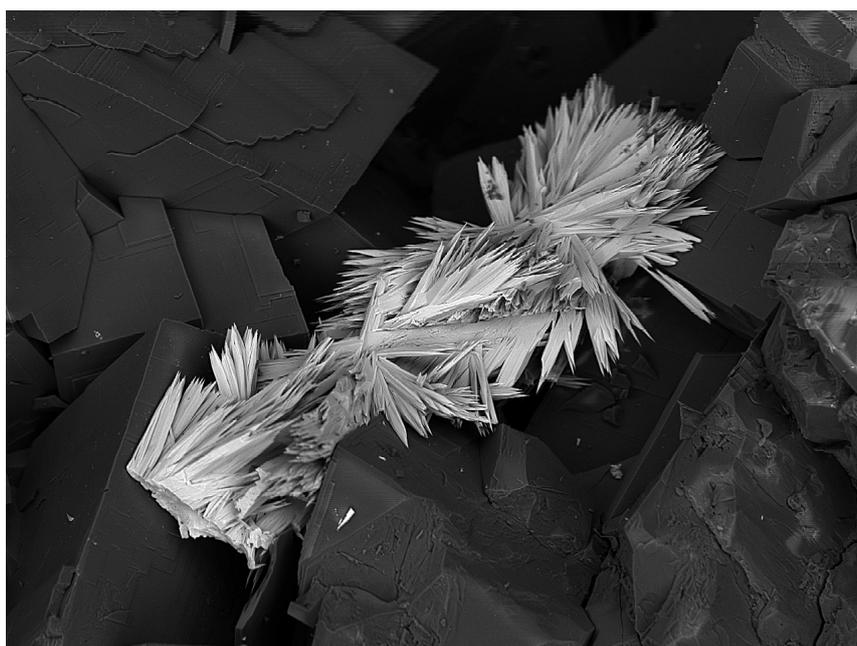


Figure 15. Aggregate of small acicular crystals of “Pb-free dalnegroite” from ribbon 1. Field of view 0.6 mm.

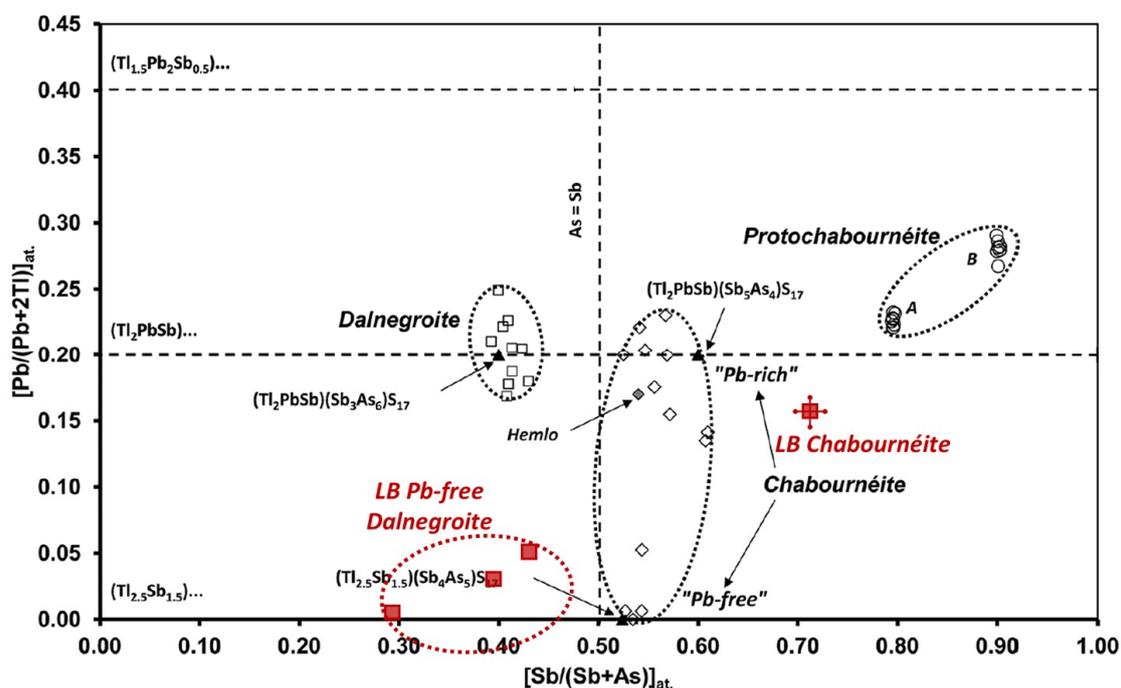


Figure 16. Diagram of Jas Roux and Abuta (France, resp. Japan, open diamonds) chabournéite, Monte Arsiccio (Italy, open circles) protochabournéite and Lengenbach (open squares) dalnegroite, modified after Reference [48], with copyright permission from Mineralogical Association of Canada, 2013. Dalnegroites are located in the As-dominated left part of the diagram, chabournéites and protochabournéites in the right part with Sb-dominance. The Pb content increases along the y-axis. The new Lengenbach samples (analyzed by EDXS) are shown as red squares. Lengenbach chabournéite is located in the Pb-rich part of the diagram (the cross gives the ± 1 sigma of all measurements), showing a higher Sb content than the samples from Jas Roux. The nearly Pb-free dalnegroite seems to be clearly isolated from the holotype material of this species (open squares).

5. Summary and Perspective

Since 1958 an uninterrupted prospection for and extraction of mineral specimens has been performed in the Lengenbach quarry. Three working collectives have been successively in charge of these purely non-profit operations, aimed at providing interesting specimens to both science and collector communities. The *Arbeitsgemeinschaft Lengenbach* (AGL, literally: Lengenbach Working Association) was active from 1958 to 1997 and extracted 28,422 specimens in total. It was followed by the *Interessengemeinschaft Lengenbach* (IGL, literally: Lengenbach Interest Association), active from 1998 to 2002. The IGL produced 2424 specimens. The *Forschungsgemeinschaft Lengenbach* (FGL), which was founded in 2003 and celebrates its 15th anniversary in 2018, has been extremely successful. The association could ensure the collaboration of several experts as Preferred Associated Scientists, who performed countless investigations to increase our knowledge of the Lengenbach mineralogy, and to contribute with many publications and the description of several new mineral species to further enhance Lengenbach's reputation as an eldorado for rare and complex sulfosalts. In these 15 years, 43 new mineral species have been described from the Lengenbach quarry, including 17 new type minerals.

As a consequence of the specimen extraction performed since 2014 in the Tl-rich dolomite ribbon 1, the number of thallium-bearing mineral samples extracted in the quarry has significantly increased recently, as it was already the case in the late eighties and early nineties when the AGL also worked in this zone (Figure 17). The FGL plans to continue the mineral prospection and extraction during the next two or three years in this fascinating part of the deposit. Consequently, there is a good chance to find additional rare thallium sulfosalts and—who knows—eventually also new mineral species.

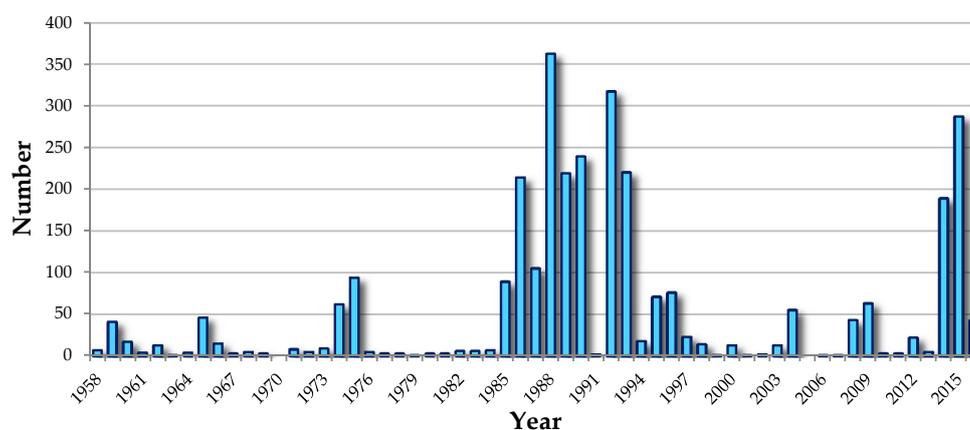


Figure 17. Number of thallium bearing samples officially cataloged by the three working associations, since 1958.

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