

## Hydrogeochemical and karstological effects of the activation of water circulation within a gypsum quarry (based on the example of Criva Quarry, Moldova Republic)

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We describe the hydrogeochemical and karstological effects of the activation of water circulation within a gypsum quarry located in the Prut River valley in the northern part of the Republic of Moldova, just next to the border with Ukraine. The quarry, located near the village of Criva, has been exploiting a gypsum deposit since 1946. Due to the almost complete filling with water of the gypsum layer, which is 25 metres thick, groundwater drainage has been carried out. As a result of the deepening of the quarry to 20 m, the upper part of the aquifer has been drained. Anthropogenic activation of karst processes are visible within the rock mass and on the surface, and since 1977, these have been observed in the labyrinthine Zolushka Cave, exposed by mining works, which is 92 km long and has a volume of 0.65 million m<sup>3</sup>. Based on the analysis of archival materials using polynomial regression analysis, the course of water drainage in the quarry was reconstructed for the years 1946–2023. In this period of 77 years, 313,003,504 m<sup>3</sup> of water were pumped out of the quarry. In order to model the course of variation of selected characteristics over this time period, a polynomial regression method based on a third-degree equation was used. This decision was dictated by the considerable fluctuations in water drainage levels and the relatively small amount of empirical data, especially in the initial and final time periods during which the study was conducted. On the basis of water chemistry, taking into account the volume weight of gypsum, the mass of gypsum drained in the dissolved state and the volume of karst voids formed were calculated. They amount to 624,435.18 tons of dissolved rock and 328,948.12 m<sup>3</sup> of newly formed karst voids, respectively. The average annual rate of chemical denudation under anthropogenic pressure was 4,272 m<sup>3</sup>/year and the rate of denudation under natural conditions was frequently exceeded by an estimated 50 to 4,000 times on an annual basis, depending on the age assessments of the karst in the area. Currently, karst in the aeration zone includes the labyrinthine system of karst voids of the Zolushka cave (which are in part filled with clayey deposits of collapsed and residual origin), and karst in the saturation zone includes the lower part of the gypsum unit with a thickness of 5 m, where new karst voids are formed. The development of karst fissures in this lower part of the gypsum, with a total volume under the cave of 165,000 m<sup>3</sup>, has caused deformation of the clay bottoms of the passages and their settlement to a depth of 2–3 m. On the surface, underground karst activation is reflected in more than 150 karst sinkholes, which began to form en masse in the quarry area after 1946. Nevertheless, the observed trend of clear degradation of the aquifer, small water inflows to the quarry and the tendency to stabilize the aquifer level are the premises for preserving the quarry and continuing its drainage after the gypsum exploitation has ended. We recommend reclaiming the quarry and creating a recreational and tourist centre on its basis.

Key words: gypsum karst, mine hydrology, data modelling, Moldova Republic.

### INTRODUCTION

We describe an example of the hydrodynamic and hydrogeochemical effects of mining in a karst area. An earlier article (Andreychouk et al., 2021) discussed these issues thematically and analysed aspects of hydrogeochemical changes

accompanying the degradation of the karst aquifer following pumping of water from the gypsum quarry in Criva, Republic of Moldova. This quarry has been in full-scale operation since 1954, although gypsum mining began at the site (an open pit of local importance) in 1946. Currently, the quarry is leased by the German company Knauf, which operates the gypsum mine.

The abundant literature on related topics includes: studies of water hazards during quarrying (Banzato et al., 2010; Caselle et al., 2020); geochemical interactions between water and rocks in karst rock quarries (Eang et al., 2018; Andreychouk et al., 2021); the activation of karst processes (Andreychouk, 2007; Van den Eeckhaut et al. 2007; Sprynsky

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et al., 2008; Rzonca and Buczyński, 2013; Byle et al., 2024); the use of quarries draining gypsum aquifers as a source of water supply (Van der Gaag, 2008); the management of quarry overburden in karst environments (Oggeri et al., 2019); and the ecological and landscape impacts of quarrying on carbonate rocks (Mayes et al., 2005; Mouflis et al., 2008; Nita, 2013).

During mining in the walls of the described quarry, a gigantic system (network) of underground voids known as the Zoloushka Cave was exposed (Andreychouk, 2007). By 2007, more than 90 km of underground passageways developed in a 24-metre thick layer of Middle Miocene (Badenian) sequence of stromatolitic (microbialitic) gypsum (Peryt, 1996) had been mapped, with a total volume of ~0.65 million m<sup>3</sup> that places the cave among the largest gypsum caves in the world (Andreychouk and Klimchouk, 2017). Geological and geomorphological studies indicate that the exposed cave labyrinth is larger and is only a part (of unconstrained size) of the extensive karst system that has developed in this gypsum deposit.

Due to its large size, the cave and its surroundings (including the quarry) have become a kind of “magnet”, attracting the attention not only of speleologists, but also of geologists, hydrogeologists, and geomorphologists, and they are of great research interest as regards the course of karst, hydrogeochemical, geomicrobiological, geological-engineering and other processes. As a result of many years of research in the region, a number of patterns have been recognised regarding the evolution of the karst water table and speleogenesis, and several interesting discoveries have been made in the fields of mineralogy and geomicrobiology (Andreychouk, 2007). As a result, both the cave and the quarry, which form a compressed mining-geological-hydrogeological system, are of great and continuing interest among a range of researchers (including those involved in environmental medicine, biology and related topics). At present, research into the region is ongoing and covers a diverse range of topics.

However, there is a danger that in the coming years the quarry, along with the cave labyrinth, will be overwhelmed by groundwater flooding. As a result, a mining facility of much scientific interest will be closed, and the unique cave system would fill with water and become inaccessible to speleologists. This situation is one reason for conducting the research we describe here. Our immediate goal was to quantify the chemical denudation of the gypsum rock mass by calculating the amount of gypsum removed in a dissolved state during quarry drainage and its karstological consequences – both in terms of the formation of new underground spaces and the appearance of karst activation at the surface. We also describe the course and scale of anthropogenically stimulated water circulation in the quarry, and compare its hydrogeochemical and karstological consequences to those resulting from natural processes that took place without human involvement.

## STUDY AREA – CRIVA GYPSUM QUARRY AND ZOLOUSHKA CAVE LABYRINTH

The quarry is located in the northern part of the Republic of Moldova, just next to the border with Ukraine and Romania (Fig. 1). The distance of the quarry from the border with Ukraine is currently just over 0.2 km, and the distance to the border with Romania, running along the Prut River, is ~1.2 km. The closest villages are Criva (on the Moldavian side) and Podvirne (on the Ukrainian side; Fig. 1).

Prior to gypsum exploitation in the late 1940s and early 1950s, both the gypsum layer and the cave itself were almost completely filled with water and were characterised by confined hydrogeological conditions and slower groundwater circulation.

Just before exposure, the hydrogeochemical environment was transitional between reducing and oxidising, with mineralization of ~2.6–2.9 g/l. Under natural conditions, water from the gypsum aquifer drained into the channel of the Prut River, located ~1 km from the quarry, and into the channel of the Pacak River, which is a left tributary of the Prut flowing near the quarry and the cave (Fig. 1).

During gradual gypsum uncovering, quarry exploitation, and associated pumping of the groundwater to the surface (at a rate of 8–25,000 m<sup>3</sup>/day), the water table was lowered, by 18 m, to the bottom of the gypsum bed. In this way, most of the gypsum layer and cave system was dewatered (Fig. 2), allowing speleologists to physically enter the cave in 1977. The cave was then mapped, and research studies and observations were conducted. Approximately 4–6 m of unexploited gypsum floor remained saturated with water (Fig. 3).

During the lowering of the groundwater table and the formation of the cone of depression, a formerly continuous karst aquifer, including the part that was visible in the cave labyrinth, started to split into individual reservoirs controlled by the morphology of different parts of the karst system, and these began to fill with clay sediments. As the water table was lowered, the aquifers shrank and more isolated mini-aquifers (cave “lakes”) formed (Andreychouk and Klimchouk, 2017; Fig. 4). In the 1950s, degradation of the aquifer led to the formation of dozens of cave “lakes” located in the lowermost parts of the cave.

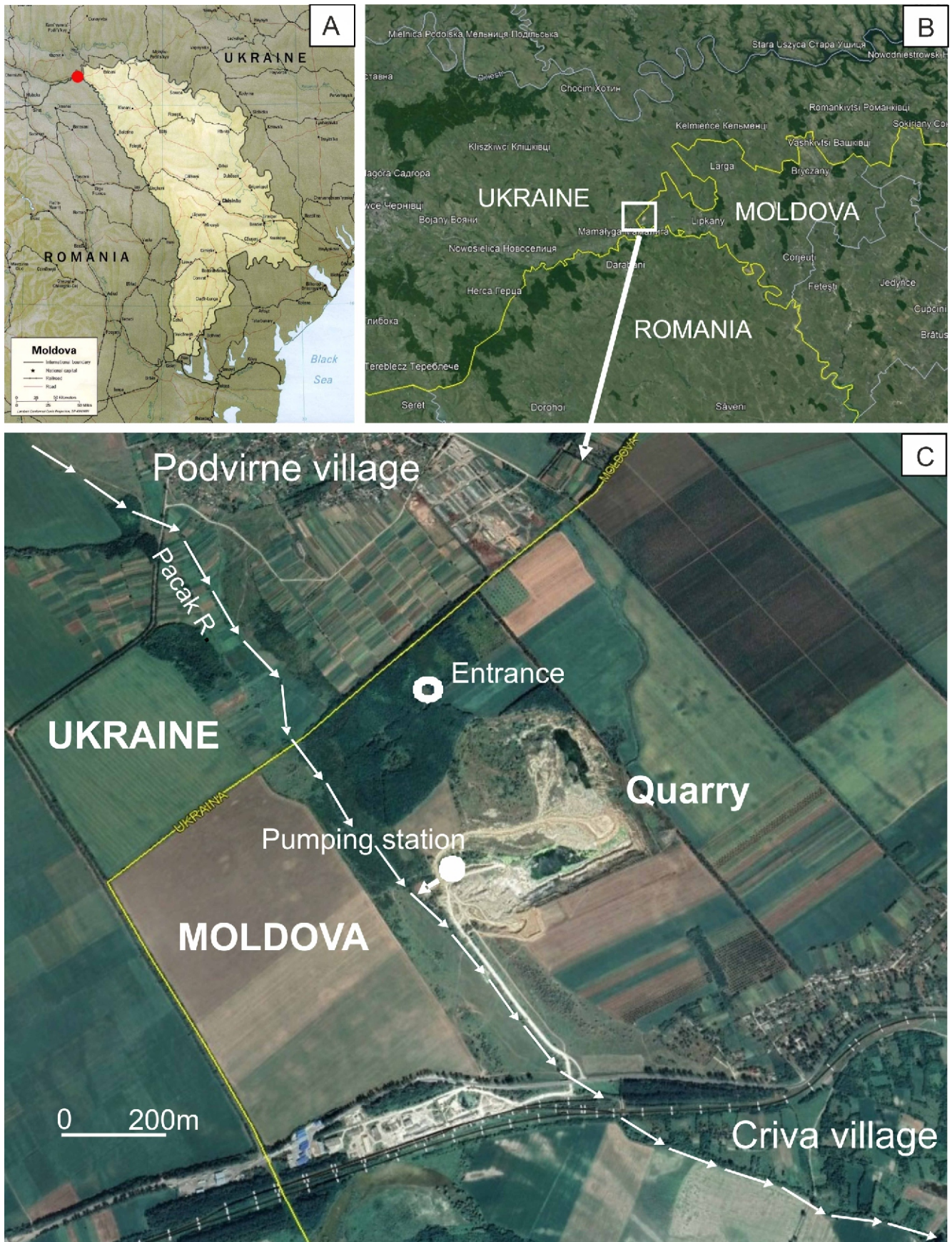
The current extraction of gypsum from the quarry takes place at two levels (Fig. 5A). The base of the lower level is water-bearing and protects against vertical seepage into the quarry of water from the lower aquifer in the Cenomanian sandstone strata (Cretaceous), which are separated from the gypsum by a layer of poorly permeable marly deposits. The water in the quarry bottom (Fig. 5A, B) represents a strongly drained aquifer in the karst gypsum. The water is discharged from the quarry (Fig. 5C) to the Pacak River bed adjoining the quarry to the south-west (Fig. 1) through a pumping station (white building in Fig. 5D). The quarry also receives (in limited amounts) water from the overlying strata, including from terrace river deposits as well as via rainfall. These issues are discussed in detail in the Methods section.

## WATER CIRCULATION WITHIN THE CRIVA QUARRY

Water inflow to the quarry and its circulation are influenced by the following factors: the lithology of the hard rock of the gypsum deposit and of the surrounding loose deposits; the hydrogeological parameters of the rocks (permeable versus poorly permeable); the strong development of karst phenomena; the nature of the compact bedrock and of the fractured rock lying directly beneath the gypsum; the blocky nature of the tectonics; the (currently absent) hydraulic relationship between the waters of the Pacak stream and the waters of the reservoir level as well as with the fractured aquifer confined under pressure that lies directly beneath the deposit; the amount of precipitation (~650 mm/year), the duration of mining (70 years), the depth of the quarry (25–45 m) and the area of the quarry (0.08 km<sup>2</sup>).

The waters circulating within the quarry and cave have diverse origins, directions, flow rates and hydrochemical parameters. They can be divided into waters flowing into the quarry from the outside (underground, from the karst aquifer and the aquifer in the terraced Quaternary deposits, and rainwater), and waters occurring (circulating) inside the cave, drained by the open-pit mine (Fig. 6).

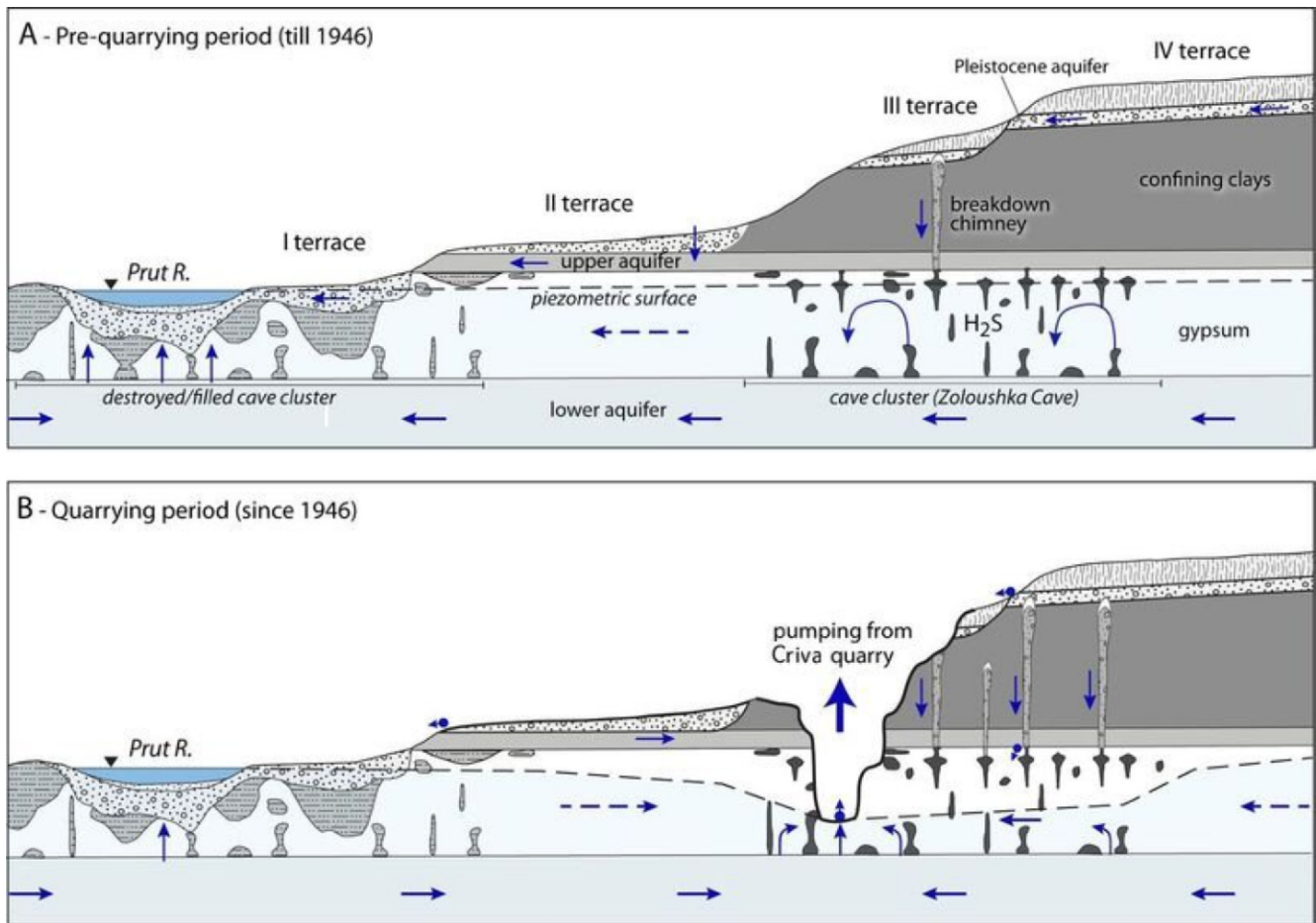




**Fig. 1.** Location of the Criva gypsum quarry on maps of the Republic of Moldova (A) and the general region (B) and the location of the quarry against the local background of Ukraine and Moldova (C), together with the neighboring villages of Criva and Podvirne (A – Internet, B, C – Google Earth)

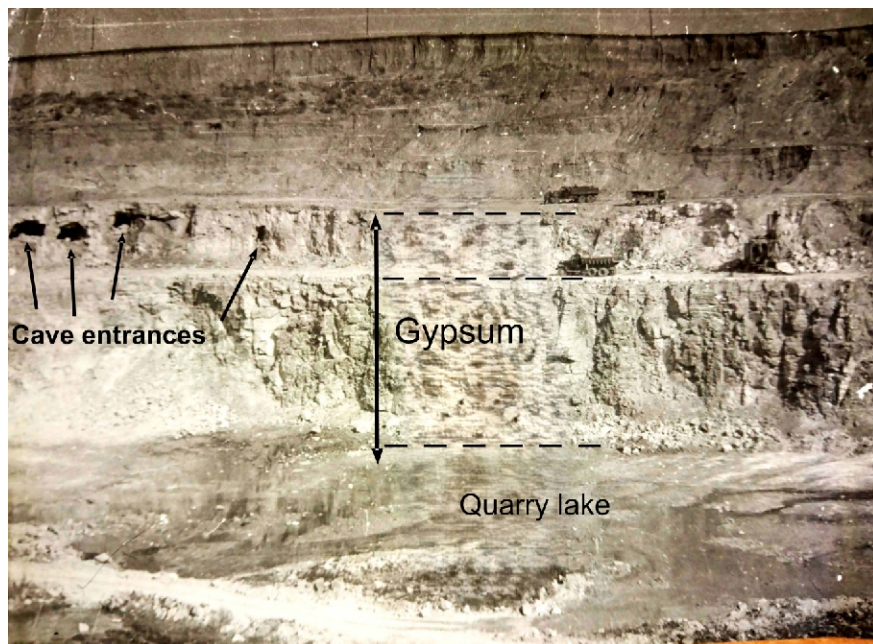
The entrance to the cave is shown in C. The yellow line shows the border between Ukraine and Moldova





**Fig. 2.** Hydrogeological conditions in the Criva gypsum quarry area in the pre-quarrying period (upper scheme) and their changes due to quarrying (lower scheme)

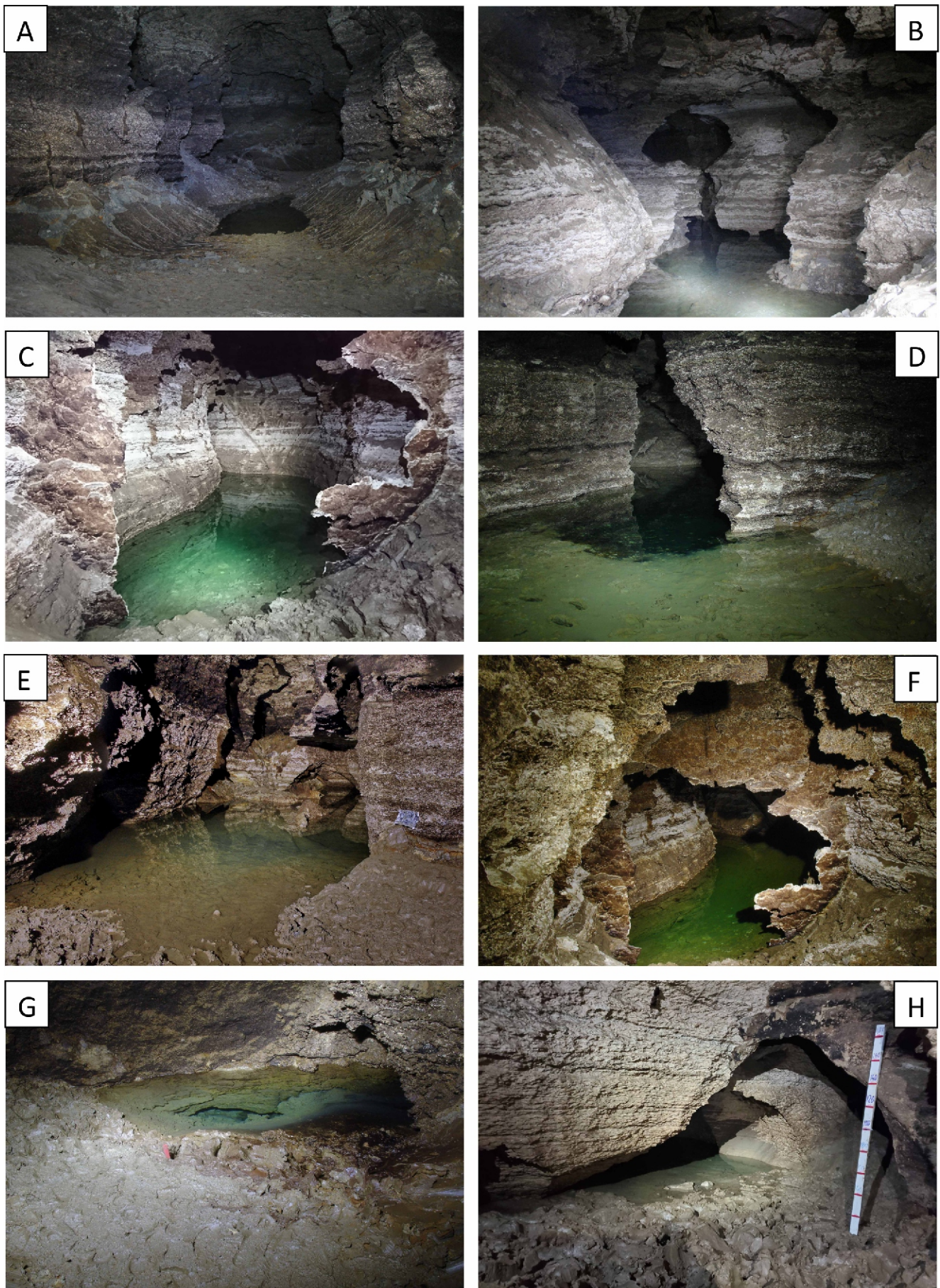
The arrows indicate the direction of ground water flow, the dotted arrows show sluggish forced flow, and the curved arrows indicate natural convection ([Andreychouk and Klimchouk, 2017](#))



**Fig. 3.** Photo of the quarry in the early 1970s

The visible part of the gypsum layer is ~18–19 m thick. The non-visible lower 4–5 m of the gypsum layer (below the dashed line) is saturated with water. The surface of the lake indicates the artificially maintained water level in the quarry. In the wall of the upper part of the gypsum layer, several entrances to the underground karst system are visible. The photo is taken from a geological report from 1975 assessing the conditions of gypsum exploitation in the quarry and its surroundings (free access to materials)





**Fig. 4. Parts of the degraded karst aquifer, with cave “lakes” visible at the bottom of cave corridors**

**A** – Fragment Lake, **B** – Vystavochnyi (Expositional) Lake, **C** – Dalekoschidne (Far-Eastern) Lake, **D** – Nautilus Lake, **E** – Krokodyla (Crocodile) Lake, **F** – Zelenyi Labirynt (Green Labyrinth) Lake, **G** – Syfon (Siphon) Lake, **H** – Filipcovo Lake (photos by O. Klimov, I. Teleshman)





**Fig. 5.** General view of the Criva Quarry (A) with a lake at the bottom (B) whose waters are directed to a pumping station (C and D; photos by V. Andreychouk)

The elements of the water circulation within the quarry - cave system (marked on Fig. 6) make different contributions to the accumulation of water on the quarry floor and its chemical composition. Some of them (the components of “internal” circulation – 2, 3, 4, 7, 8, 9, 10, 11, 12, 13, 14 – see Fig. 6) participate indirectly and their importance is negligible and difficult to quantify. The following components of the balance should be considered in more detail: the flow into the quarry of groundwater (from the karst aquifer and sub-gypsum waters), precipitation (rain and snow) and the seepage of deep water from unconsolidated Quaternary deposits.

The main component of the water flowing into the quarry is groundwater. Of secondary importance are rainwater and seepage water from the Quaternary overburden. The amount of precipitation in the study area, according to data from meteorological stations in Lipkani (Moldova) and Botoșani (Romania), varies between 520 and 790 mm/year and averages ~650 mm. The quarry recharge area is  $0.2 \times 0.4$  km, or  $0.08 \text{ km}^2$ , hence the total atmospheric water supply is  $52,000 \text{ m}^3$  per year.

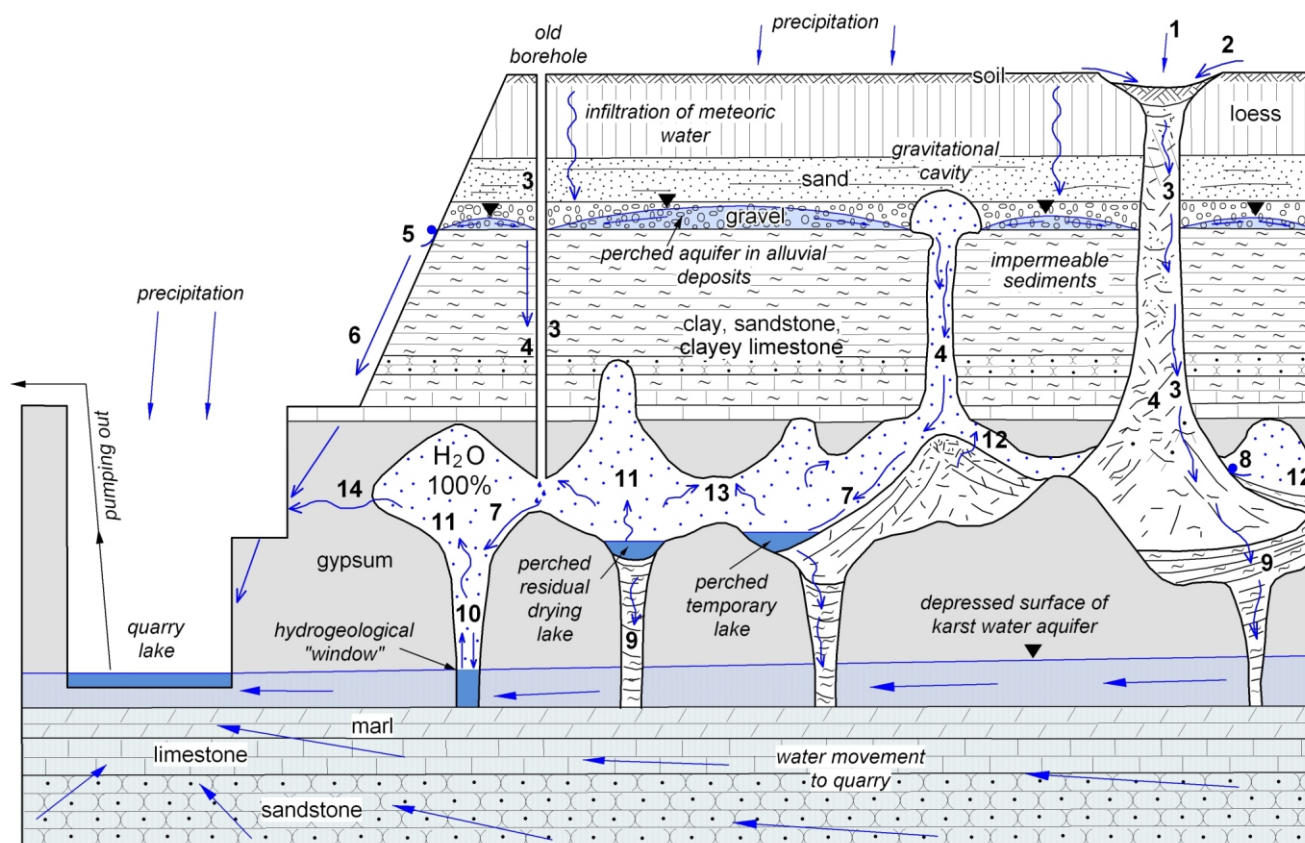
The water supply from the aquifer in the Quaternary deposits is difficult to estimate. Some of the water, accumulating in supra-gypsum terrace deposits, undergoes underground drainage, infiltrating through sinkhole chimneys filled with loose deposits (accumulating during the development of the sinkhole chimneys towards the surface). In the area occupied by the cave ( $\sim 1.0 \text{ km}^2$ ), several instances of the seepage of water from the collapse cone were observed. The largest seepage

was in the form of a spring with a capacity of  $\sim 0.05 \text{ l/s}$  (the so-called “Spring of Life” in the Chamber of Chernivtsi Caverns). This spring functioned for several years and disappeared as a result of disturbance of the collapse material. If we assume that, within the cave boundaries, the inflow of infiltration water (including hidden water) is  $0.05\text{--}0.5 \text{ l/s}$ , the total amount of water entering the cave can be estimated at  $1,500\text{--}15,000 \text{ m}^3$  per year.

The surface drainage is definitely greater. At many points along the walls of the quarry, where terraced sandy-gravel deposits were exposed lying on poorly permeable Baden (supra-gypsum) clay strata, we observed 1–2 outflows of water, combining into streams flowing down to the quarry. The output of such springs usually ranges from  $0.1$  to  $0.3 \text{ l/s}$  and is highly variable depending on atmospheric conditions. In total, the amount of water seeping from the overburden into the quarry may be  $3,000\text{--}9,000 \text{ m}^3$  per year for outflows with a capacity of  $0.1 \text{ l/s}$  and  $9,000\text{--}27,000 \text{ m}^3$  for outflows with a capacity of  $0.3 \text{ l/s}$ . In total,  $\sim 10,000\text{--}40,000 \text{ m}^3$  of water per year flows into the quarry from the underground and surface drainage of the Quaternary level.

The estimated values quoted above are small compared to groundwater inflows from the karst aquifer of several thousand  $\text{m}^3$  per day (see below). On an annual basis, precipitation and water from Quaternary formations account for only  $0.1\text{--}0.3\%$  of the total amount of water discharged from the quarry. Due to the small inflow and low mineralization ( $580 \text{ mg/l}$  on average) of





**Fig. 6. Circulating water within the gypsum massif under conditions of mining activity**

1 – precipitation, 2 – surface runoff, 3 – water in terrace deposits above the gypsum, 4 – water infiltrating into the cave]] via collapse tubes and boreholes, 5 – outflows of water from Quaternary deposits in the quarry wall, 6 – runoff of water from Quaternary deposits into the quarry, 7 – seepage of infiltration waters into cave “lakes”, 8 – outflow of infiltration water in the form of small springs, 9 and 10 – infiltration of water into cave deposits and lakes, 11 and 12 – evaporation of waters from the surface of cave lakes and deposits in the bottoms of passages, 13 – accumulation of water vapour in cave air, 14 – natural drainage of moisture from the cave into the quarry (through fissures)

Quaternary-derived waters and waters from precipitation, their contribution was ignored in calculations of the chemical denudation within the quarry.

## MATERIALS AND METHODS

In order to evaluate the chemical denudation, as expressed in the load of dissolved substances in water pumped out of the quarry, and its karstological consequences, it was necessary to obtain data on:

1. The amount of groundwater flowing into and out of the quarry.
2. The chemical composition of the pumped-out water.
3. Information on exogenous processes occurring within the quarry and in its vicinity, especially the formation of new sinkholes, which can be considered a consequence of the anthropogenic activation of water circulation in the karst rock mass.

In relation to (1), it is not possible to obtain detailed and regular data on the pumping of water from the quarry for the entire period of its operation. Data from the period 1946–1991, in the form of pumping logs and energy consumption, have been lost. Later, after the political transition, pumping observations were carried out irregularly. It was only after the German Knauf

Group took over the mining plant that the amount of water pumped out of the quarry began to be documented consistently. We were able to obtain detailed data for a period of 9 years (2003–2011; [Table 1](#)).

Regarding the earlier years (1946–2002), and the later period (2012–2024), selected data found in geological reports and a few archival documents were used. In the Discussion, we attempt to estimate the missing values based on some factual and theoretical premises. The data available to us served as base values for the preparation of a multi-year curve (model) describing the amount of pumped water and the resulting hydrochemical calculations of denudation and the formation of underground voids in the “quarry-cave” system.

For (2), the chemical composition of water from the gypsum aquifer is provided on the basis of our previous studies. Specifically it is derived from hydrogeochemical studies of the water from the quarry’s crevasse system ([Andreychouk, 2007](#)) and the water circulating in the Zoloushka Cave system, which is drained by the quarry ([Andreychouk, 2007](#); [Andreychouk et al., 2009, 2021](#)). The results of these studies are very consistent, hence averaged values of constituent concentrations in water pumped from the quarry were adopted for chemical denudation studies. The average concentrations of selected components in the water, studied in 2020, were: TDS 2.646 g/l, Ca 0.543 g/l,  $\text{SO}_4$  1.439 g/l.

Table 1

Groundwater drainage at Criva gypsum quarry, Moldova in 2003–2011 (m<sup>3</sup>)

Year/ month	2003		2004		2005		2006		2007	
	month	day	month	day	month	day	month	day	month	day
01	253,770	8,186	265,898	8,577	208,879	6,738	280,688	9,054	311,968	10,063
02	256,990	9,178	243,151	8,385	255,417	9,122	280,203	10,007	277,402	9,907
03	267,981	8,645	222,228	7,169	239,400	7,723	294,000	9,484	313,120	10,101
04	261,500	8,717	230,301	7,677	231,000	7,700	284,128	9,471	231,000	7,700
05	254,243	8,201	190,168	6,134	188,013	6,065	289,002	9,323	212,100	6,842
06	244,360	8,145	209,748	6,992	229,530	7,651	286,440	9,548	214,200	7,140
07	269,598	8,697	207,629	6,698	219,139	7,069	274,514	8,855	157,000	5,065
08	223,522	7,210	226,800	7,316	255,150	8,231	270,900	8,739	165,000	5,323
09	240,834	8,028	250,904	8,363	264,857	8,829	271,320	9,044	132,000	4,400
10	219,866	7,092	193,007	6,226	252,000	8,129	248,466	8,015	169,000	5,452
11	208,230	6,941	235,444	7,848	274,207	9,140	293,712	9,790	173,000	5,767
12	227,949	7,353	224,129	7,230	288,628	9,311	313,320	10,107	166,000	5,355
Annual total/daily average	2,928,843	8,024	2,699,407	7,375	2,906,220	7,962	3,386,693	9,279	2,521,790	6,909
Year/ month	2008		2009		2010		2011			
	month	day	month	day	month	day	month	day		
01	204,395	6,593	357,157	11,521	277,407	8,949	460,303	14,848		
02	166,828	5,753	300,920	10,747	231,193	8,257	401,722	14,347		
03	168,000	5,419	352,130	11,359	255,679	8,248	380,035	12,259		
04	231,000	7,700	275,533	9,184	252,373	8,412	291,736	9,725		
05	138,600	4,471	343,270	11,073	240,236	7,750	353,940	11,417		
06	142,810	4,760	270,000	9,000	265,020	8,834	287,727	9,591		
07	205,800	6,639	280,132	9,037	265,020	8,549	272,704	8,797		
08	419,513	13,533	267, 880	8,641	503,506	16,242	271,271	8,751		
09	276,448	9,215	256,200	8,540	347,430	11,581	216,859	7,229		
10	240,167	7,747	282,790	9,122	293,996	9,484	265,719	8,572		
11	231,725	7,724	237,300	7,910	238,533	7,951	303,804	10,127		
12	357,464	11,531	126,470	4,080	295,510	9,533	376,950	12,160		
Annual total/daily average	2,782,750	7,603	3,349,782	9,177	3,465,903	9,496	3,882,770	10,638		

With respect to (3), information on the exogenous processes occurring in and around the quarry is incomplete, as there was no monitoring of sinkhole formation during the study period. Each newly formed sinkhole was backfilled with material from the overburden and its place of formation was not recorded on mining maps. Andreychouk (2007), who has been studying the Zoloushka cave region since 1976, documented tens of newly formed sinkholes. Because the quarry has been in operation since the 1950s, the total number of sinkholes created in that period is significantly higher than before mining activity started. Hence, the description of the effects of surface karst activation governed by mining activities is mainly qualitative. The development of subterranean karst, on the other hand, has been documented quantitatively (see Discussion section).

Geological and exploration reports found in the archives of the geological institutions of Chernivtsi, Kyiv and Chisinau proved to be very helpful as sources of data from the early periods of quarrying. These include reports from 1946, 1975, 1976 and 1991 (see References).

Following the acquisition of selected data on the quantities studied, i.e., water drainage, CaSO<sub>4</sub> content and volume of newly formed karst voids in the gypsum rock mass, regression models based on a third-degree polynomial function were de-

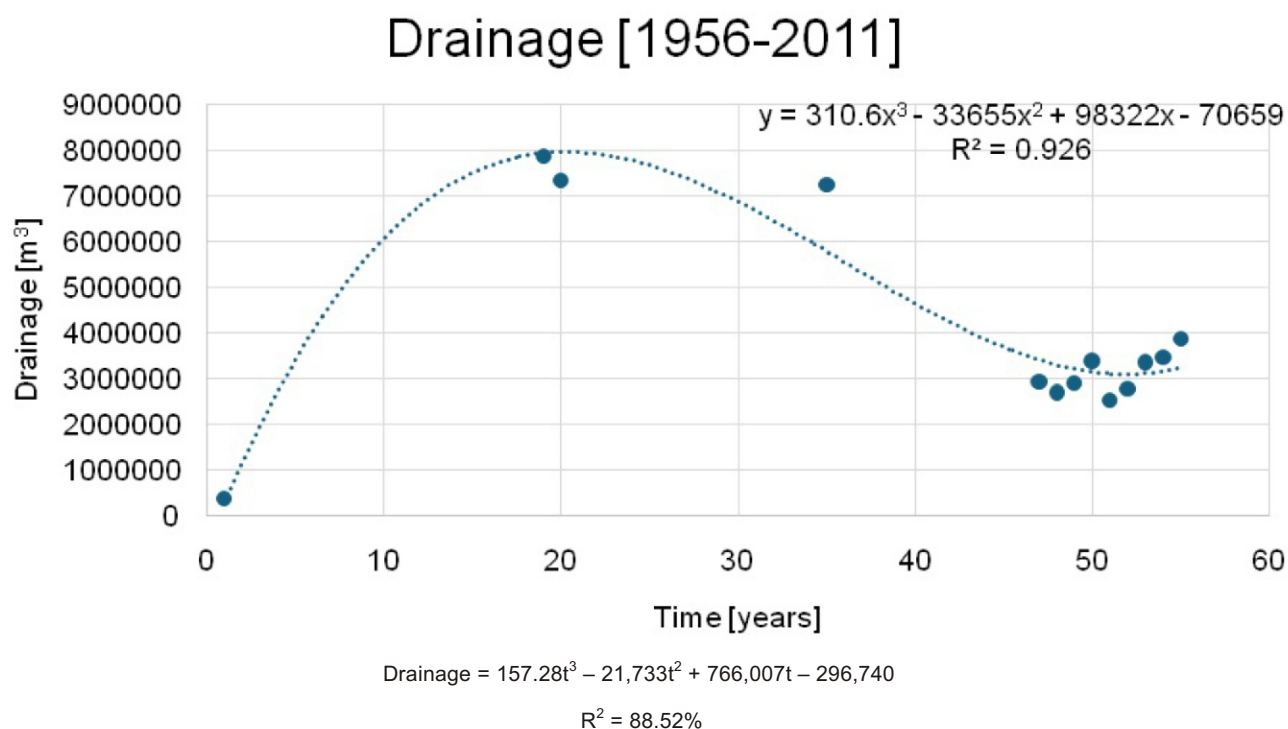
veloped. This type of function accurately captures the variability of the results, where large fluctuations can be observed, which is difficult to reflect with traditionally used functions (Morton and Henderson, 2008; Keshtegar et al., 2019; Inforsato and De Jong van Lier, 2021). Because the goal was to obtain a unified forecasting model, and not to have too many results for different time intervals, a model based on folding functions of different types for different sub-periods was abandoned. Similar methods were also used for the other hydrological characteristics (Kelleners and Chaudhry, 1998; Elgama et al., 2021; Zawawi et al., 2022).

## RESULTS

The more or less regular acquisition of regular data, the results of many years of research, as well as interpolations made on the basis of this data, allowed us to reconstruct the course of the pumping of water from the quarry (Fig. 7), and to carry out calculations of the amount of dissolved material removed from the rock mass (chemical denudation) in the period 1956–2023 (Figs. 8, 9 and Tables 2–4).

Based on the data, a third-degree polynomial regression was derived with the general equation:





Dependent variable: drainage – year scale [m <sup>3</sup> ]; confidence level: 95.0% (alfa = 0.050)						
	Rating	Standard error	Value t df = 10	p	Lower confidence limit	Upper confidence limit
a	157	35.0	4.49590	0.001150	79	235
b	–21,733	3,734.4	–5.81950	0.000168	–30,053	–13,412
c	766,007	115,408.7	6.63734	0.000058	508,860	1,023,153
d	–296,740	980,516.4	–0.30264	0.768368	–2,481,467	1,887,987

Fig. 7. Variability of the amount of water pumped out of the Criva gypsum quarry for the period 1956–2023

$$y = at^3 + bt^2 + ct + d$$

For the variable “drainage”, the model shown in Figure 7 was obtained.

The quarry water drainage values predicted from the model are shown in Table 2.

When considering the amount of chemical denudation, it is necessary to take into account the amount of water circulating in the aquifer system, the hydrochemical data and the geochemical composition of the gypsum rocks. The calculation assumes that the average bulk weight of gypsum is 2.32 t/m<sup>3</sup>. The bulk gravity of gypsum from the Criva Quarry was measured in 1975 at the quarry laboratory using 4 samples. 2 samples of coarse crystalline gypsum had volumetric weights of 2.29 t/m<sup>3</sup> and 2.33 t/m<sup>3</sup>, and 2 samples of fine gypsum had volumetric weights of 2.31 t/m<sup>3</sup> and 2.34 t/m<sup>3</sup>. Averaging the results from the 4 samples gives a value of 2.32 t/m<sup>3</sup>.

The average content of the main component in the gypsum of the deposit exploited – CaSO<sub>4</sub> · 2H<sub>2</sub>O – is 97.38%. The insoluble and barely soluble residue is 2.62%. The main impurity in the gypsum rock is CaCO<sub>3</sub> (0.3–2.0%). The content of SiO<sub>2</sub> varies from 0.10 to 0.80%, that of Fe<sub>2</sub>O<sub>3</sub> from 0.02 to 0.09%, and that of MgO from 0.0 to 0.5%. The content of impurities in the

vertical profile of the gypsum unit varies. The gypsum showed high chemical purity throughout the profile (Andreychouk, 2007).

For the load of CaSO<sub>4</sub> removed from the quarry with the pumped water, the model shown in Figure 8 was obtained.

The values of CaSO<sub>4</sub> load removed with pumped water predicted from the model are shown in Table 3.

For the “space equivalent” variable, the model shown in Figure 9 was obtained.

The “space equivalent” values predicted from the model are shown in Table 4.

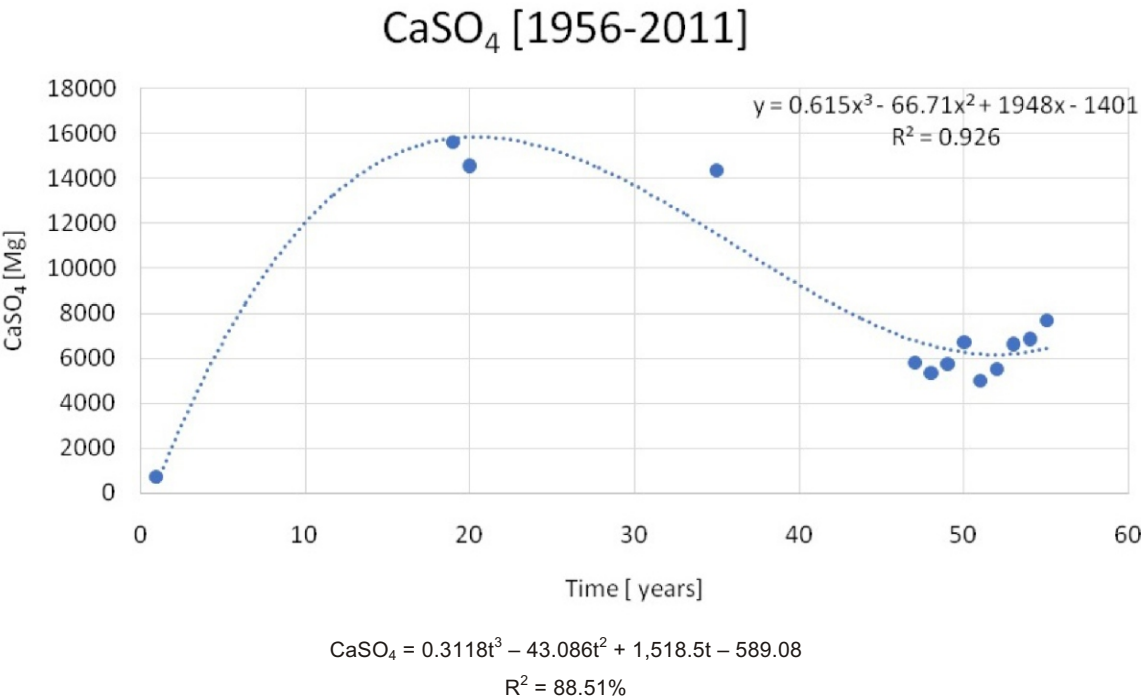
The fitted polynomial model is of good quality, as corroborated by the high value of the R<sup>2</sup> coefficient of determination, which is ~89%. This means that the model reflects the natural variability of the estimated variables with corresponding accuracy, so 11% remains a random component. The high quality of the model parameters is also indicated by their significance levels. All parameters of the model turned out to be statistically significant at the 5% level. The significance value for free expression is statistically less relevant.

The choice of the polynomial model stemmed from the estimated random variables that showed changes depending on the period considered. One can see a clear increase in the val-

Table 2

Predicted annual average values of water pumped from the Criva gypsum quarry in the period 1957–2023

Year	Drainage [m³]	Year	Drainage [m³]	Year	Drainage [m³]
1957	447,691.28	1980	7,743,458.72	2003	4,026,673.44
1958	1,149,600.24	1981	7,727,810.00	2004	3,792,673.76
1959	1,809,930.56	1982	7,692,287.28	2005	3,560,504.72
1960	2,429,625.92	1983	7,637,834.24	2006	3,331,110.00
1961	3,009,630.00	1984	7,565,394.56	2007	3,105,433.28
1962	3,550,886.48	1985	7,475,911.92	2008	2,884,418.24
1963	4,054,339.04	1986	7,370,330.00	2009	2,669,008.56
1964	4,520,931.36	1987	7,249,592.48	2010	2,460,147.92
1965	4,951,607.12	1988	7,114,643.04	2011	2,258,780.00
1966	5,347,310.00	1989	6,966,425.36	2012	2,065,848.48
1967	5,708,983.68	1990	6,805,883.12	2013	1,882,297.04
1968	6,037,571.84	1991	6,633,960.00	2014	1,709,069.36
1969	6,334,018.16	1992	6,451,599.68	2015	1,547,109.12
1970	6,599,266.32	1993	6,259,745.84	2016	1,397,360.00
1971	6,834,260.00	1994	6,059,342.16	2017	1,260,765.68
1972	7,039,942.88	1995	5,851,332.32	2018	1,138,269.84
1973	7,217,258.64	1996	5,636,660.00	2019	1,030,816.16
1974	7,367,150.96	1997	5,416,268.88	2020	939,348.32
1975	7,490,563.52	1998	5,191,102.64	2021	864,810.00
1976	7,588,440.00	1999	4,962,104.96	2022	808,144.88
1977	7,661,724.08	2000	4,730,219.52	2023	770,296.64
1978	7,711,359.44	2001	4,496,390.00		
1979	7,738,289.76	2002	4,261,560.08		



Model: $v17=a*v1^3+b*v1^2+c*v1+d$ (Data - year scale, dependent variable: CaSO <sub>4</sub> [kg] Confidence level: 95.0% (alfa = 0.050)						
	Rating	Standard error	Value t df = 10	p	Lower confidence limit	Upper confidence limit
a	0.312	0.069	4.49569	0.001151	0.16	0.466
b	−43.086	7.405	−5.81855	0.000169	−59.59	−26.587
c	1,518.495	228.843	6.63554	0.000058	1,008.60	2,028.389
d	−589.081	1,944.257	−0.30299	0.768110	−4,921.15	3,742.992

Fig. 8. Variability in the amount of the CaSO<sub>4</sub> load removed with pumped water from the Criva Quarry for the period 1956–2023

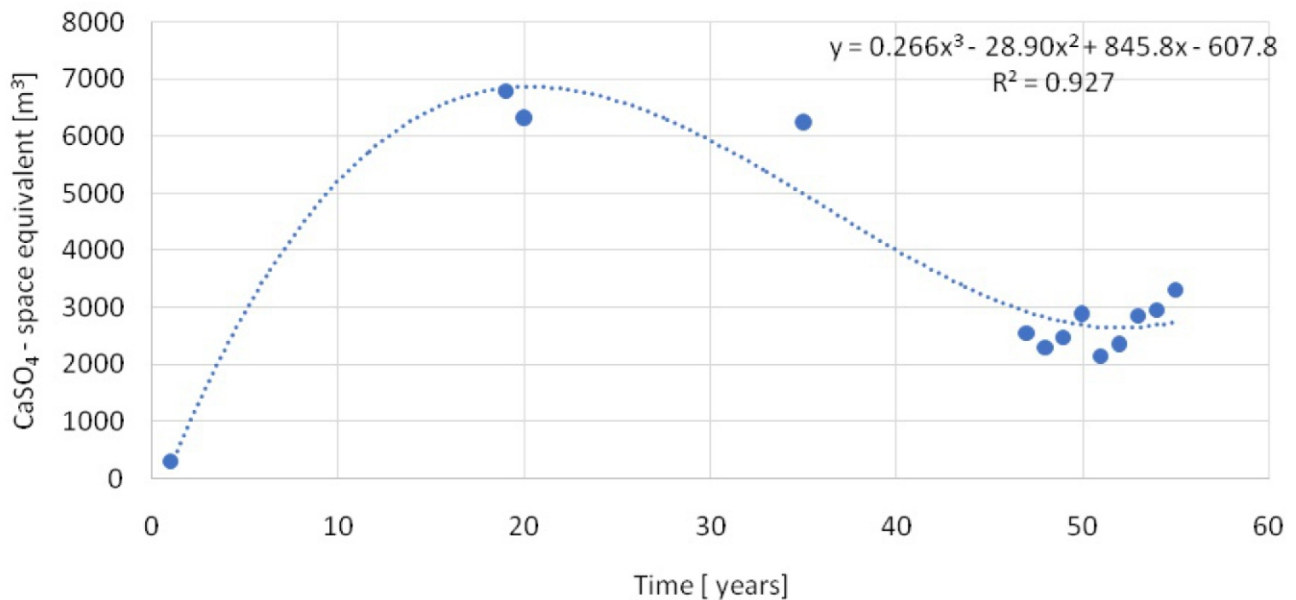


Table 3

Predicted annual values of removed  $\text{CaSO}_4$  load with water pumped from the Criva gypsum quarry in the period 1957–2023

Year	$\text{CaSO}_4$ [Mg]	Year	$\text{CaSO}_4$ [Mg]	Year	$\text{CaSO}_4$ [Mg]
1957	886.6458	1980	15,347.71	2003	7,975.457
1958	2,278.07	1981	15,316.55	2004	7,511.362
1959	3,587.065	1982	15,245.98	2005	7,050.892
1960	4,815.499	1983	15,137.89	2006	6,595.92
1961	5,965.245	1984	14,994.13	2007	6,148.316
1962	7,038.173	1985	14,816.58	2008	5,709.95
1963	8,036.153	1986	14,607.12	2009	5,282.695
1964	8,961.058	1987	14,367.61	2010	4,868.419
1965	9,814.756	1988	14,099.92	2011	4,468.995
1966	10,599.12	1989	13,805.92	2012	4,086.293
1967	11,316.02	1990	13,487.49	2013	3,722.183
1968	11,967.33	1991	13,146.5	2014	3,378.538
1969	12,554.91	1992	12,784.8	2015	3,057.226
1970	13,080.64	1993	12,404.29	2016	2,760.12
1971	13,546.4	1994	12,006.83	2017	2,489.09
1972	13,954.04	1995	11,594.28	2018	2,246.006
1973	14,305.44	1996	11,168.52	2019	2,032.741
1974	14,602.47	1997	10,731.42	2020	1,851.163
1975	14,847.01	1998	10,284.85	2021	1,703.145
1976	15,040.92	1999	9,830.689	2022	1,590.557
1977	15,186.07	2000	9,370.795	2023	1,515.269
1978	15,284.34	2001	8,907.045		
1979	15,337.6	2002	8,441.309		

### $\text{CaSO}_4$ - space equivalent [1956-2011]



	Model: $v_3 = a \cdot v_5^3 + b \cdot v_5^2 + c \cdot v_5 + d$ dependent variable: $\text{CaSO}_4$ – space equivalent [ $\text{m}^3$ ] Confidence level: 95.0% (alpha = 0.050)					
	Rating	Standard error	Value. t df = 9	p	Lower confidence limit	Upper confidence limit
a	0.266	0.0516	5.15543	0.000599	0.15	0.383
b	–28.902	4.3508	–6.64285	0.000095	–38.74	–19.060
c	845.831	100.1506	8.44559	0.000014	619.27	1,072.387
d	–607.864	655.5867	–0.92721	0.378014	–2,090.90	875.176

Fig. 9. Variability in the volume of newly formed karst voids in the gypsum rock mass for the period 1956–2023

Table 4

**Predicted values of volumes of newly formed karst voids in the gypsum rock mass for the period 1957–2023**

Year	Space equivalent [m <sup>3</sup> ]	Year	Space equivalent [m <sup>3</sup> ]	Year	Space equivalent [m <sup>3</sup> ]
1957	382.35	1980	6,675.75	2003	3,450.96
1958	988.83	1981	6,661.50	2004	3,248.59
1959	1,559.33	1982	6,630.07	2005	3,047.85
1960	2,094.66	1983	6,582.28	2006	2,849.56
1961	2,595.65	1984	6,518.96	2007	2,654.54
1962	3,063.11	1985	6,440.91	2008	2,463.61
1963	3,497.84	1986	6,348.96	2009	2,277.59
1964	3,900.69	1987	6,243.92	2010	2,097.28
1965	4,272.45	1988	6,126.62	2011	1,923.52
1966	4,613.96	1989	5,997.86	2012	1,757.12
1967	4,926.03	1990	5,858.48	2013	1,598.90
1968	5,209.47	1991	5,709.27	2014	1,449.67
1969	5,465.10	1992	5,551.07	2015	1,310.25
1970	5,693.75	1993	5,384.69	2016	1,181.46
1971	5,896.22	1994	5,210.95	2017	1,064.12
1972	6,073.34	1995	5,030.67	2018	959.05
1973	6,225.93	1996	4,844.66	2019	867.06
1974	6,354.80	1997	4,653.74	2020	788.97
1975	6,460.77	1998	4,458.73	2021	725.60
1976	6,544.66	1999	4,260.45	2022	677.76
1977	6,607.28	2000	4,059.72	2023	646.29
1978	6,649.46	2001	3,857.35		
1979	6,672.01	2002	3,654.16		

ues of the variables in the initial period, while there was a decrease in the 1990s. After that, a slight increase can be observed again, and in the last period a clear decrease can be observed. Thus, the variability has a tendency that can be colloquially described as a “sine wave” of variable amplitude, and this kind of relationship can be most easily approximated by polynomial equations. The accuracy of the model selection is confirmed by measures of its quality of fit, which allows reliable assessments of all variables estimated.

The models and calculations generated show that during the 67-year period of operation considered, a total of 313,003,504 m<sup>3</sup> of water was pumped out of the quarry. Along with the karst waters pumped out of the quarry, 620,947.5 tons of gypsum rock was removed, which is equivalent to 269,548.21 m<sup>3</sup> of newly formed karst voids.

However, at this point we should make some correction regarding the last value (volume). In the above calculations, we took into account the content of only the CaSO<sub>4</sub> component in the gypsum rock, ignoring the volume of the 2 water molecules that gypsum contains as mineral, i.e. we counted as if it was the anhydrous form anhydrite. However, these two related minerals have different densities due to the presence of water in gypsum. In the case of gypsum from the deposit described here, it is 2.32 g/cm<sup>3</sup> (2.32 Mg/m<sup>3</sup>), while in the case of anhydrite it is 2.95 g/cm<sup>3</sup> (2.95 Mg/m<sup>3</sup>). The difference is 0.63 g/cm<sup>3</sup> (21.36%). This is the amount of underestimation (relative to gypsum) of the volume of rock per 2 water molecules in the gypsum formula. Therefore, it is necessary to add it to the total value of the volume of voids formed during the study period.

With its contribution (269,548.21 m<sup>3</sup> + 21.36% of 269,548.21 m<sup>3</sup> i.e. 57,575.50 m<sup>3</sup>), the total volume of voids formed is estimated at 327,123,71 m<sup>3</sup>.

## DISCUSSION

### HYDROGEOLOGICAL CHARACTERISTICS OF THE GYPSUM DEPOSIT AND WATER INFLOWS

Mine hydrogeology classifies deposits according to the complexity of the hydrogeological conditions. Deposits in Poland are divided into those with simple and complicated hydrogeological conditions (Dowgiało et al., 2002). The gypsum deposit in Criva may be assigned to the category of deposits with simple hydrogeological conditions based on its poor water-bearing capacity (inflow to the mine of less than 600 m<sup>3</sup>/h). However, it may also be assigned to the category of deposits with complicated hydrogeological conditions due to its water-bearing capacity in compact rock with developed karst features. The criteria for classifying open-pit mined deposits into individual degrees of water hazard are included in the (Ordinance of the Minister of the Environment, 2021) based on natural hazards in mining plants. Large-scale open-pit mines are vulnerable to increased inflows of water due to the possibility of exceeding the capacity of the drainage system, with hazards posed by groundwater flowing into the pits, surface watercourses, and rainwater associated with prolonged or sudden and heavy rainfall.



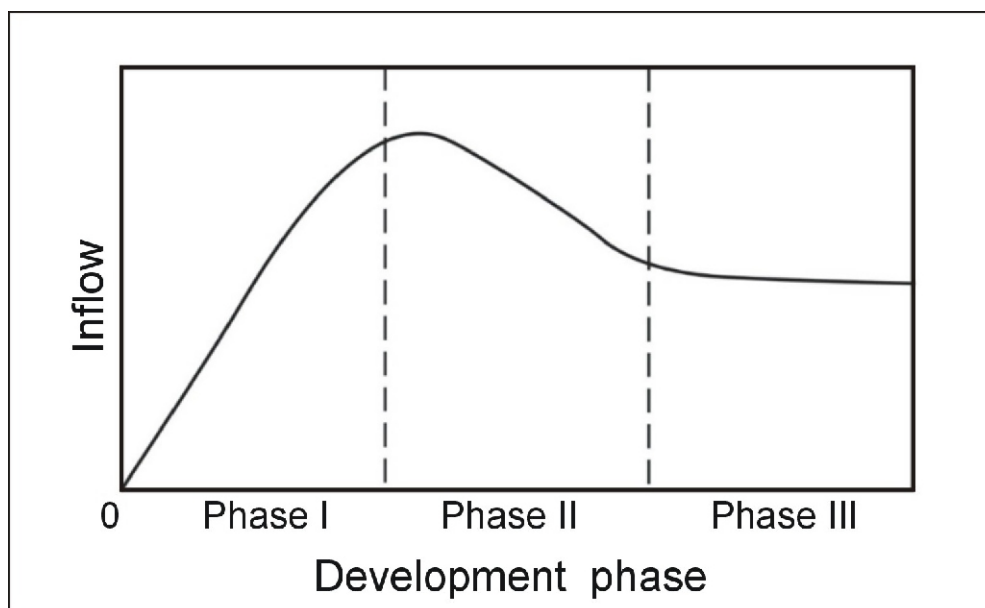


Fig. 10. General curve of development of mine water inflows (Wilk, ed., 2003)

The Criva deposit has a relatively high degree of water hazard. Although intense precipitation has never directly contributed to flooding in the quarry, the associated hydrological system clearly responds to precipitation. On several occasions, water from the Pacak River has broken into the quarry pit through karst funnels in the riverbed, causing disappearing of the river. Water flowing from the slopes or bottom of the open pit, however, poses little danger to its operation.

The formation of mine water inflows is influenced by the following factors: time, mine depth, mine area, and mining of the deposit as a function of the increase in mined area (Rogoz, 2004). The practice of open pit mining shows that as the deposit is exploited, accompanied by active drainage of inflowing groundwater, regular changes occur in the inflow of water to the pits. Three phases of tributary development can be distinguished (Fig. 10).

In Phase I, intense depletion of static resources in the vicinity of access pits usually takes place, forming a narrow and deep depression funnel. Inflows increase rapidly, and inflow from dynamic resources is small. In Phase II, there is a gradual decrease in inflows due to flattening and widening of the funnel of depression, a small increase in the area of mine workings, and a gradual increase in the proportion of inflows from dynamic resources. In Phase III, the expansion of the funnel of depression is proportional to the increase in the area of mine workings, inflow from static resources is approximately constant, inflow from dynamic resources may increase slightly, and there is stabilization and progressive reduction of inflows.

The pattern of depletion of karst aquifer resources established for the Criva gypsum deposit (Fig. 7) generally repeats the general trend shown in Figure 10. A comparison of the two figures shows that the aquifer drained by the Criva Quarry is currently in the third phase of development of mine water inflows.

#### WATER DRAINAGE DURING THE QUARRY OPERATION PERIOD

The models presented above and the resulting tables can be used to estimate chemical denudation over a period of 67 years (1957–2023), for which irregular but reliable measurements were available. The calculation of quarry water drainage and chemical denudation for 1946–1956 is shown below.

According to the 1975 geological report, the quarry's gypsum began to be exploited on an industrial scale only in 1954. In 1946–1948 there was only a small pit, where raw material was extracted from the upper reliable part of the gypsum with a thickness of 3–5 m. A pump with a capacity of 3 m<sup>3</sup>/hr (72 m<sup>3</sup>/day) was installed in the casting pit, which was unable to pump out the inflowing water, given the heavy rainfall.

In 1947–1948, geological and technical works were carried out to examine the exploitation of the deposit and to assess its resources. In 1948–1953, the quarry was deepened, new drainage pumps were installed and the drainage of groundwater increased. In the period 1948–1954, the inflow of water to the quarry increased progressively. In 1957, it reached 1,000 m<sup>3</sup>/day (with a range of fluctuations of 750–1,250 m<sup>3</sup>/day). Based on the general trend (Fig. 7), it can be assumed that in 1954, at the start of the industrial exploitation of raw materials, the pumping capacity could have already amounted to 750 m<sup>3</sup> per day (minimum value in 1957). During the preparatory mining works, in 1948–1954, the pumping level increased from 72 m<sup>3</sup>/day to 750 m<sup>3</sup>/day. Accordingly, it can be assumed that from 1954 to 1957, the drainage of water from the quarry increased from 750 to 1,000 m<sup>3</sup> per day. Table 5 shows the inferred volume of water drainage, corresponding to the inferred drainage of dissolved gypsum and the volume of newly created underground voids.

Based on Table 5, chemical denudation in the period 1946–1956 can be estimated at 3,487.68 tons of gypsum discharged in a dissolved state, which corresponds to a spatial equivalent of 1,503.31 m<sup>3</sup>. In total, in the period 1946–2023, chemical denudation was estimated at 624,435.18 tons of gypsum discharged in dissolved state, which corresponds to 271,051.52 m<sup>3</sup> of newly formed karst spaces. We correct this value by 21.36% (57,896.60 m<sup>3</sup> - underestimated volume due to the presence of bound water in the gypsum - see above) and obtain a corrected total of 328,948.12 m<sup>3</sup>.

Considering that after the dissolution of each m<sup>3</sup> of gypsum rock in the karst system, ~0.06 Mg of insoluble residue (2.62%) remains, it should be assumed that with the significant increase in the volume of the karst system, ~16,263 tons of carbonate-clay terrigenous sediments were deposited within it, which corresponds to a volume of ~8,130 m<sup>3</sup>. At the same time, however, some carbonates were dissolved, hence the amount of the residual sediment may be smaller.

Table 5

**Documented and estimated amounts of water discharged from the quarry and the accompanying chemical denudation in the period 1946–1956**

Year	1946	1947	1948	1949	1950	1951	1952	1953	1954	1955	1956	1957
Drainage [m <sup>3</sup> /day]	72	72	72	~ 200	~ 310	~ 420	~ 530	~ 640	~750	~835	~920	1000
Drainage [m <sup>3</sup> /year]	26,280	26,280	26,280	73,000	113,150	153,300	193,450	233,600	273,750	304,775	335,800	
CaSO <sub>4</sub> load [Mg] per year	52.09	52.09	52.09	144.69	224.26	303.84	383.42	463.00	542.57	604.06	665.56	
Chemical denudation [m <sup>3</sup> ] per year	22.45	22.45	22.45	62.37	96.66	130.97	165.27	199.57	233.87	260.37	286.88	

#### CHEMICAL DENUDATION RANGE

It is interesting to compare the rate of chemical denudation in the study area in natural conditions and in the time of human influence. The gypsum cave in the study area began to develop in the Middle Pleistocene (Andreychouk, 2007). This was associated with the erosive dissection of the Badenian–Cenomanian aquifer system following deposition of the third terrace of the Prut River. It is estimated that the water circulation in this aquifer system was initiated at that time, comprising ascent of water from the underlying marls and sandstones through the fractured gypsum unit towards the layer of fractured limestone above the gypsum, within which these waters were transported to the higher part of the Prut. This means that the formation of the karst system with a volume of 650,000 m<sup>3</sup> took ~700,000 years. The average rate of underground chemical denudation was ~0.9 m<sup>3</sup>/year within the cave field, whose area was 2.5 km<sup>2</sup>. The average rate of denudation for the period of anthropogenic activation of the karst aquifer (1946–2023) for the cave area is 4,272 m<sup>3</sup>/year, assuming that the total volume of voids formed over the 77-year period was 328,948.12 m<sup>3</sup>. This means that the rate of chemical denudation in anthropogenically activated conditions was almost 4,000 times higher than that occurring through natural karst processes (Andreychouk, 2007).

This resulted from the significant activation of water circulation in the aquifer system drained by the quarry and from the high solubility of gypsum rocks. For comparison: in the case of carbonate karst development in natural and anthropogenically modified mining conditions, the geological and geomorphological effects would be smaller (the solubility would be one order of magnitude lower; Rózkowski and Rózkowski, 2014). In the case of salt karst, however, due to the very high solubility of salt (two orders of magnitude higher than that of sulphate rocks), the effects of anthropogenic karst activation take on a catastrophic character in a very short period of time (Andreychouk, 1996, 2002; Molenda and Kidawa, 2022; Molenda et al., 2023).

The development of the underground karst was not uniform, but took place exponentially. The labyrinth system developed more rapidly with the increase in volume, the increase in the amount of circulating water and the increase in the water-rock contact surface. The currently observed size of the cave corridors is the result of several thousand years of development, during which the Prut riverbed began to cut into the gypsum ceiling, which facilitated the upwards drainage of the karst aquifer, leading to the activation of water circulation and the acceleration of karstification. If we assume that the entire volume of the cave system is the result of the activity during the Holocene Epoch, then the average rate of natural chemical de-

nodation in the karst system (65 m<sup>3</sup>/year) turns out to be about 66 times lower than denudation in anthropogenic conditions, calculated for the last 77 years (4,272 m<sup>3</sup>/year).

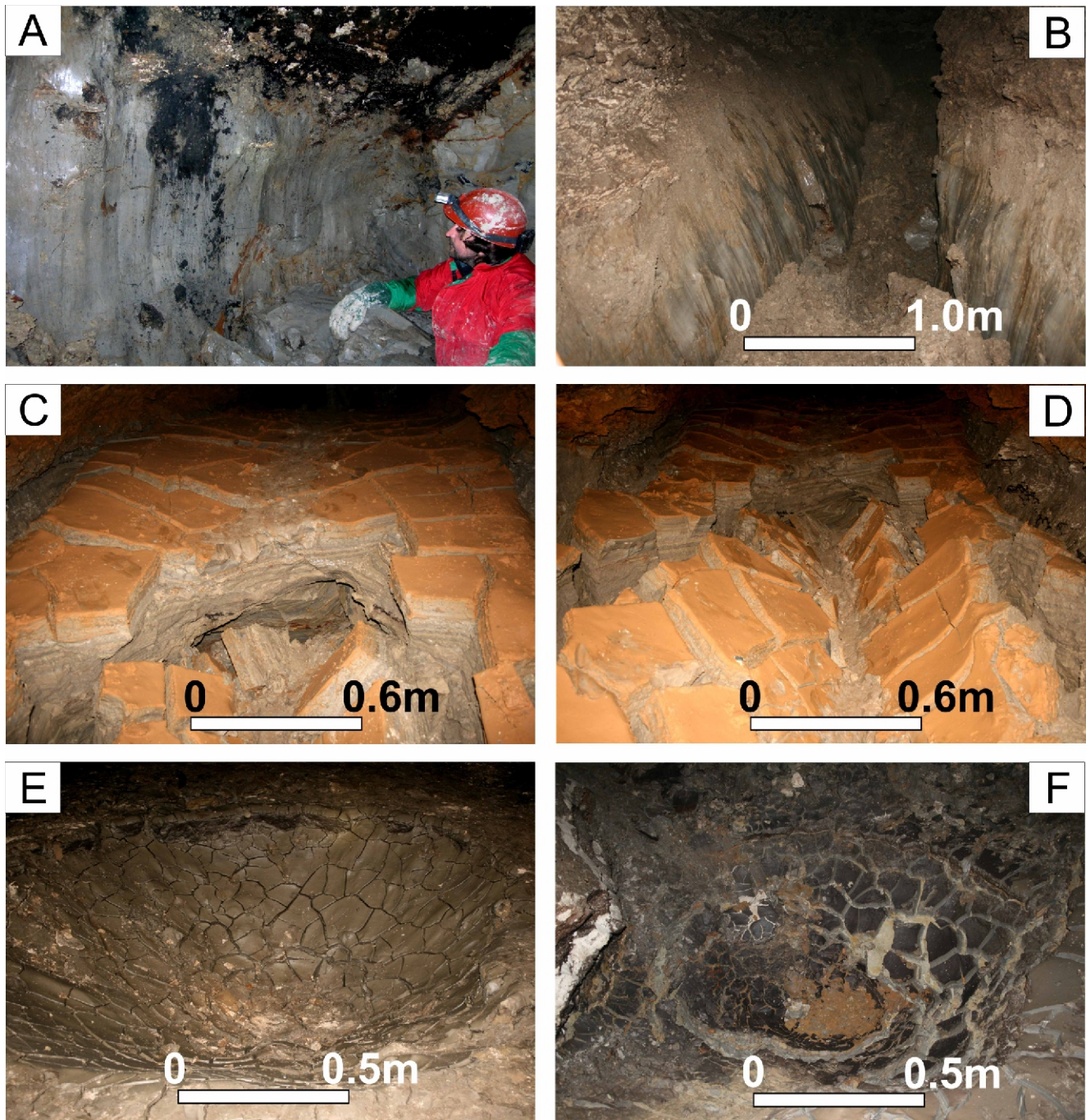
The quoted amount of chemical denudation during the period of anthropogenic activation of water circulation is lower than previously inferred. The first attempt to assess the extent of denudation can be found in the calculations given in Voropay et al. (1985), which showed a rate of 6,500 m<sup>3</sup>/year. This number is about one and half times as high as that resulting from our calculations. This is because we made calculations based on the water pumping reaching its maximum efficiency at that time (see Fig. 7 and Table 2). A lower value of denudation (4,861 m<sup>3</sup>/year) was inferred by Andreychouk (2007), whose calculations also referred to the maximum drainage efficiency, but who did not have data for the entire period.

#### SPATIAL VARIABILITY OF DENUDATION

Gypsum dissolution processes occur under anthropogenic influence in the lower, water-filled part of the gypsum layer with a thickness of 4–6 m. It is here that new anthropogenically conditioned voids were created. Spaces created in natural conditions in the middle and upper part of the gypsum profile are located higher, in the aeration zone. Now let us consider the relationship between the newly created space and the total volume of the water-bearing layer, i.e. its current degree of karstification. According to Andreychouk (2007), the area of the quarry's influence, within which karst processes have been significantly activated, is at least 5 km<sup>2</sup>, hence with an average thickness of water-filling zone (5 m), we obtain a maximum volume of the aquifer of 25 million m<sup>3</sup>. Beyond the influence of mining drainage, as one moves away from the quarry, the thickness of the water-bearing layer gradually increases. Comparing the calculated volume of new rock voids (328,948.12 m<sup>3</sup>) and the volume of the aquifer within the range of the quarry's impact (25 million m<sup>3</sup>), we obtain a volumetric karstification index of the rock mass of 1.3%. Outside the range of the quarry's impact, it will be significantly lower.

In the lower part of the gypsum layer, during the period of the quarry's operation, empty space has developed at a rate of ~4,272 m<sup>3</sup>/year. The area of the cave surface currently explored is ~2.5 km<sup>2</sup>, which is half the area of the estimated range of the quarry's impact, corresponding to half the volume of rock voids (164,474.06 m<sup>3</sup>). Assuming part of the void space is filled with residual sediment (in the amount of 8,130 m<sup>3</sup> / 2 = 4,065 m<sup>3</sup>), we obtain a figure of 164,469.99 m<sup>3</sup> of empty space in the water-saturated gypsum zone under the cave. What is the





**Fig. 11. Selected examples of cave corridor bottom subsidence above the widening, corroded cracks at their bottom (photos by V. Andreychouk)**

relationship between the newly formed voids in the lower part of the aquifer and the voids in the aeration zone, covered by the cave? The interactions between them are reflected in the subsidence in the clay deposits filling the bottoms of the cave corridors. The processes of deformation of the corridor bottoms are continuous phenomena and have a diverse character (Fig. 11).

Bottom deformation structures cover the entire cave. Their intensity and nature (size, shape, rate of formation) are locally variable in the region and depend on local factors – the morphology of the cave corridor, its width, the degree of filling with cave deposits, etc. Within wide corridors, there is bottom subsidence across their entire width (Fig. 11A, B); within large cave chambers, there is mainly the formation of craters (Figs. 11E, F

and 12A). A characteristic feature of bottom deformation in areas with smaller corridors is the uneven, complex nature of deformation along the corridors – intact segments change suddenly or stepwise into canyons several metres deep (Fig. 11A). The depth of subsidence ranges from 0.5 to over 3.0 m. In the areas of the cave where there are wide corridors, subsidence occurs commonly, while in the areas where the corridors are smaller, subsidence is of a non-uniform nature, as shown in Figure 11B (the larger deformation structures are marked on the plan). The situation shown in Figure 12B represents the state of the cave in 1986. Currently the scale and nature of subsidence are greater and more diverse.



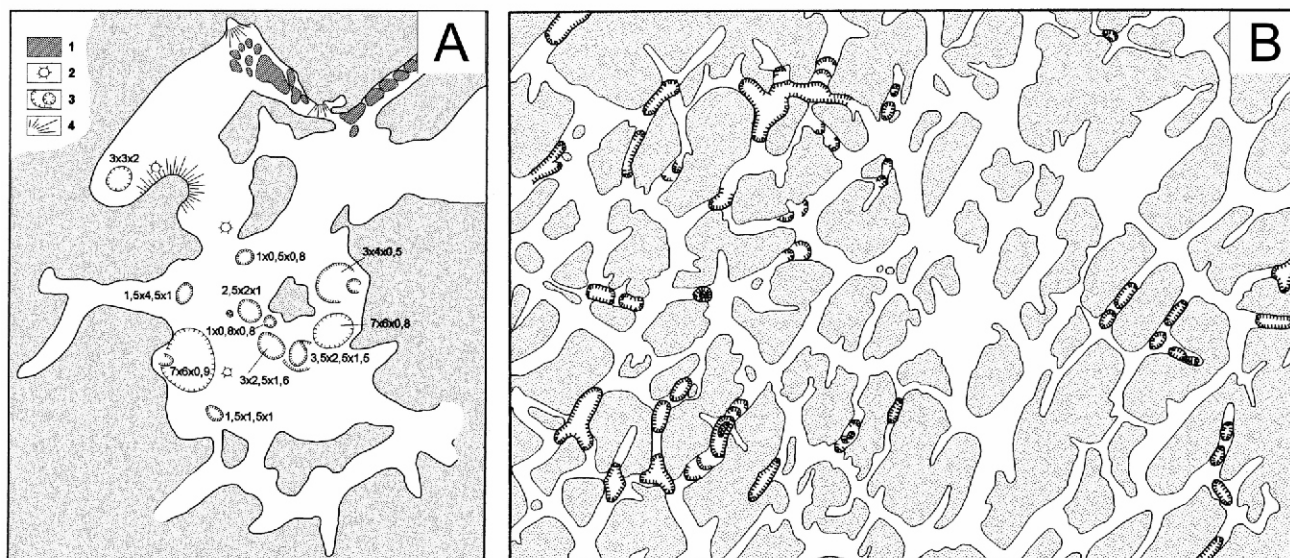


Fig. 12. Examples of deformation in the bottom of corridors as of 1986 (Andreychouk, 2007)

**A** – subsidence and sinkhole forms in the Ancient Hall: 1 – collapses, 2 – pile cones, 3 – sinkhole forms and subsidence troughs, 4 – slopes of sinkhole cones; **B** – subsidence on the bottoms of corridors filled with clay deposits in the Wesely area

#### COURSE OF DENUDATION IN THE SHORT TERM

The regime used to pump water from the quarry is determined by both natural factors (precipitation, water inflow) and technical factors (pump efficiency, pump failures, etc.). Natural factors are mostly cyclical (seasonal); however, during the course of the quarry's activity, catastrophic events have occurred, such as flooding as a result of the intrusion of water from the Pacak River infiltrating through karst sinkholes formed in its bed. The rapid increase in the intensity of groundwater inflow to the quarry was partially caused by floods on the Prut River, with the river level rising by 5–9 m, and intense regional rainfall. Accidental factors included drainage pump failures.

The processes shaping water inflow to the quarry are complex, as seen in the course of climatic and hydrological episodes in the period 05–07.1976 shown in Figure 13 (based on the geological report from 1976). A number of episodes can be distinguished, revealing the rapid response (1–2 days) of the karst aquifer in the gypsum and the Pacak River to increased rainfall recharge and the hydraulic bond between the drained aquifer and the Pacak River, as well as water escapes into the karstified Pacak River bed at increased flow rates in the river:

1. 22–25/05/1976 rainfall of 15 mm (22–23/05), combined with an increase in water drainage ( $Q$ ) in the quarry (from an average of 820 to a maximum of 910  $m^3/h$  on May 23), an increase in flow rate ( $Q$ ) in the Pacak River (22–25/05 with 130 to a maximum of 410  $m^3/h$  on May 24);
2. 28–29/05 – rainfall 8 mm (28/05), then an increase in the flow rate ( $Q$ ) in the Pacak River (29–30/05 from 170 to a maximum of 250  $m^3/h$  on 29/05);
3. 03–13/06 – consequence: rainfall (up to 18 mm 03–04/06), a sharp increase in the flow rate in the Pacak River from 120  $m^3/h$  on 03/06 to a maximum of 1,020  $m^3/h$  on June 4 and a drop to 0  $m^3/h$  on 06/06 – then water escapes into sinkholes in the Pacak River (06–07/06), a decrease in water drainage ( $Q$ ) from the quarry, probably as a result of pump failures (from 875  $m^3/h$  on June 4 to 400  $m^3/h$  on June 5, and then a

sharp increase in drainage water from the quarry to 1450  $m^3/h$  on 05/06/06 and further decrease to 1,070  $m^3/h$  on 08/09/06).

4. 14–17/06 – increase in drainage of water from the quarry from 1,080  $m^3/h$  to 1,440  $m^3/h$ . combined with light rainfall Wed. 2–5 mm, preceded by water escapes from the Pacak River (12–13/06), which continued on 15–17/06, with increased water inflows to the quarry of 1,150–1,200  $m^3/h$ ;
5. 20/06 – break in quarry drainage from 9 a.m. to 11 a.m. on June 20, 1976 (pump failure) and then on June 21–22, a sudden increase in quarry drainage from 715  $m^3/h$  to 1,340  $m^3/h$  (June 21) (probable partial flooding of the quarry);
6. 27–29/06 – increase in quarry drainage from 910 to 1,410  $m^3/h$  (28/06); correlations with water escapes from the Pacak River (27–29/06 and 30/06);
7. during the rainless period 02–07/07 there was a decrease in the drainage of water from the quarry from 1,115 to 875  $m^3/h$  and minimum flow rates in the Pacak River of 10–30  $m^3/h$ , which documents the clear dominance of underground flows in the karst during the rainless period;
8. 08–12/07 – an increase in rainfall lasting 5 days to 48 mm on 10/07 and then an increase in the flow rate in the Pacak River (9–15/07) from ~40  $m^3/h$ , exceeding 100  $m^3/h$  (with maximum on 12/07 – 420  $m^3/h$ ) in the period 10–14/07; at the same time water inflows to the quarry show a decreasing tendency from 1,015  $m^3/h$  on 08/07 to 865  $m^3/h$  on 15/07/1976.

#### KARST ACTIVATION ON THE SURFACE

The reflections of underground denudation and karst at the ground surface are sinkhole forms (Fig. 14). Due to the lack of monitoring of these phenomena, we have only limited data. In the case of sinkhole formation, mining services in the quarry eliminated sinkholes by filling them with material from heaps of overburden of the gypsum deposit.



**Fig. 13. Episodes revealing response of the gypsum karst aquifer and the Pacak River to increased rainfall recharge (1976)**

The authors' many years of field observations allow us to draw the following conclusions regarding newly formed sinkholes.

1. The formation of new sinkholes is related to the beginning of deposit exploitation and the lowering of the karst water level. The number of sinkholes formed in the period 1954–2023 is estimated at over 150. This number is comparable to the total number of sinkhole-type karst forms mapped and described at the ground surface before exploitation began, which was 140.
2. The new sinkholes mostly relate to the surface of the flood terrace of the Pacak River and the Prut River and to the surface of the I terrace of the Prut River. They often form in groups of several individual forms. Locally they also occur at the foot of the II terrace of the Prut (Fig. 14A), but only very sporadically within the higher terraces (II, II and III). In the case of the high terraces, they are located at the bottom of pre-existing karst forms (as re-activation of karst forms).
3. Small forms with a diameter of 2–5 m and a depth of 1–5 m dominate among the newly formed sinkholes. The shapes of the sinkholes formed within the flood terrace and ter-

race I are irregular (Fig. 14B), while the sinkholes from terrace II and higher terraces are circular (Fig. 14D) and are usually deeper.

#### IMPLICATIONS AND APPLICATION OF RESEARCH RESULTS

Our research allows a number of useful conclusions, that can be considered in scientific (theoretical), methodological and practical perspectives.

From a scientific point of view, we show, using a specific example, how significant the influence of an anthropogenic mining facility, such as a quarry, can be on the course of denudation processes. They also describe the dynamics of these processes within the context of the long-term evolution of the karst aquifer, i.e. its degradation under the influence of groundwater pumping. The trends established, showing degradation of the aquifer and stabilization of its level, along with a radical decrease in the amount of pumped water, show a clear decrease in underground denudation and of the rate of formation of new voids. This fact opens up some new possibilities regarding the





**Fig. 14. Some examples of new karst sinkholes appearing sporadically within the Criva Quarry area**

**A, B** – sinkholes on the slope (A) and floodplain (B) of the Pacak River valley (a left tributary of the Prut River); **C** – sinkhole on the border passage between Ukraine and Moldova situated 1.0 km from the quarry; **D** – sinkhole just near the railway with fresh clay infills (0.8 km from the quarry) (photos by V. Andreychouk)

future fate of the quarry and cave (see below). The calculated values of denudation can serve various purposes, both scientific and practical. The developmental patterns established undoubtedly are of more than local scope and may have universal aspects, constituting valuable comparative material for other researchers dealing with similar problems.

The methodology described can be efficiently used to monitor the variability of selected features through time. In our study there was a limited amount of data available to estimate drainage,  $\text{CaSO}_4$  contents and their equivalent, making it difficult to obtain a reliable simulation. The selection of polynomial regression was not random. Other typical functions used for regression, like linear, exponential, logarithmic or hyperbolic ones were not good enough to show the empirical course of the data through time. This kind of function can be used efficiently, especially in cases where significant and often quite random changes occur (Ostertagová, 2012). This allows taking into consideration all the characteristic points and local amplitudes through the entire period of time analysed. However, having more data, especially from the period between 1957–2000 would certainly allow better constraint of the function obtained, not only for the known values but also for the predicted ones. Such simulation is a source of potential process explanation, and may be used in various datasets where significant amounts of data are missing.

The highly active dissolution processes and their consequences in the form of actively developing deformation of the cave corridor floors and the formation of sinkholes on the ground surface should be taken into account in the scope of further exploitation of the quarry and use of the surrounding areas, as well as of the cave itself. While the established progressive degradation of the karst aquifer creates favourable conditions for the continuation of gypsum extraction (lower water pumping, lower energy costs, etc.), the activation of karst and the formation of new voids in the gypsum contribute to the serious degradation of the cave, which has been under legal protection since 1981, and the degradation of the areas around the quarry. It also creates a serious geological and engineering threat (sinkhole formation) for existing buildings in the villages of Podvirne and Criva and for transport infrastructure, both road and rail. Comprehensive monitoring of the processes and phenomena taking place is needed, both at the surface and in the cave, and reliable documentation of the volume of water pumped out of the quarry. Particular attention should be paid to the south-eastern wall of the quarry (from the Prut River side), where large water inflows, including river inflows (from the Prut River bed), may occur, which threaten to cause a mining disaster, with flooding of the quarry and cave. The Pacak River bed should also be subject to periodic inspection. The formation of sinkholes at its bottom or in the immediate vicinity (usually they develop in

stages – see Andreychouk 1991, 2007) poses a risk of the stream disappearing and water ingress, which may cause the quarry to flood. Such events have already been observed in the past, during quarry exploitation.

Among these implications, one cannot ignore the research into the cave, which is an object of exceptional scientific and potentially practical importance. For several decades, the cave has been a unique research ground for speleologists, mineralogists, geologists, hydrogeologists, engineering geologists, geophysicists, geochemists, biologists and microbiologists, medical researchers and other specialists. Several tens of scientific articles, two monographs and several chapters in monographic studies, have been published on the cave. Nevertheless, there remain many unsolved research problems, and so it is important to continue multi-faceted scientific research into the cave.

The existence of the cave in a form accessible to speleologists and scientists is, however, dependent on the exploitation of gypsum in the quarry and the pumping out of groundwater. The observed trend of clear degradation of the aquifer (Fig. 7), small water inflows to the quarry (Table 2) and the tendency to stabilise the aquifer level (Figs. 7 and 10) are reasons for preserving the quarry and continuing its drainage after gypsum exploitation has finished. We advise reclaiming the quarry and creating a recreational and tourist centre on its basis. Recreational services may use the cave by creation of a tourist route, peripheral areas with a high (2–5%) CO<sub>2</sub> content may be used for medical and speleological purposes, with organization of underground monitoring points integrated into an externally managed system. In addition, the quarry itself would become very attractive object, with a lake at the bottom, a swimming area, viewing platforms, a geological/speleological museum, and service points. The location of the object at the border of 3 countries, the high population density of in the area, good transport accessibility (close to national road and railway lines connecting Lviv, Kishinev and Odessa) would undoubtedly ensure great tourist popularity of this object.

Voropay et al. (1985) showed that the karst waters pumped out of the quarry have balneological properties and can be used for this purpose. Hydrogeochemical studies indicate a high content of therapeutically active ions in the water, with concentrations higher than in sulphate waters in famous health resorts such as Krainka and Nizhneivkino (Russia), Likenaj (Lithuania) or Baldone (Latvia). The waters from the Criva Quarry do not contain toxic compounds, are not polluted – it is an agricultural and forested area, with isolation of the gypsum by overlying clay-rich deposits), are characterized by low (< 1 mg/l) contents of organic compounds, high mineralization and high concentra-

tions of alkaline earth metals. Due to these properties, these waters have choleric, relaxing and anti-inflammatory properties, which qualifies them for the treatment of gastric problems, gastric and duodenal ulcers, chronic colitis, other intestinal diseases, gout, kidney stones, and chronic inflammatory processes of the liver and gallbladder.

## CONCLUSIONS

Opencast exploitation of the Criva gypsum deposit has become a significant factor in the activation of local karst, which is developing dynamically in the gypsum unit. The effects of this activation include an increase in the volume of underground voids developed in the gypsum and the formation of new sinkhole forms in the areas adjacent to the quarry. The rate of development of both underground voids and sinkholes significantly exceeds the rate of karst development prior to the commencement of gypsum deposit exploitation (respectively: 3,520 m<sup>3</sup>/year and 65 m<sup>3</sup>/year). The main factor accelerating the development of karst is mining activity – specifically, the pumping of water from the quarry – which has significantly increased the dynamics of water flow in the karst system, dissolving gypsum and removing substances dissolved in water from the rock mass.

We analysed the data via polynomial regression based on a third-degree function. The methodology applied effectively reflects the irregular nature of variability for the quantities studied, this being corroborated by the satisfactory level of the determination coefficient R<sup>2</sup> and the statistical significance of the equation parameters at the 0.05 level. Thus, the models developed can be considered a source of reliable forecasts for the years for which empirical data are not available and can be used to predict the level of the quantities studied in subsequent years.

The patterns of karst development shown, accelerated by mining activity, are known from many other karst areas. The uniqueness of our research lies in the presence of one of the largest accessible cave systems in the world, Zoloushka, within the exploited gypsum deposit, allowing for the observation of the course of karst processes and other physicochemical processes within the cave system.

The quantitative and morphological effects of karst development (both in terms of the size of underground voids and the number of karst forms at the surface) for 700,000 years in natural conditions and for only 70 years in anthropogenically modified conditions are comparable (in relation to the resulting voids: 650,000 m<sup>3</sup> and 328,948.12 m<sup>3</sup>, respectively).

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