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Abstract

Meghalaya (NW India) is one of the regions in the world with highest recorded rainfalls. Because of these peculiar climatic conditions and intense solutional weathering, karstic caves are widely reported in the numerous limestone areas of this part of India. Likewise, the extremely high rainfall and the tropical monsoon climate have fostered the formation of several caves in quartz-rich lithologies. The most important of these caves is Krem Puri, over 24 km long, characterized by a fracture-controlled labyrinth forming a bi-dimensional maze with mostly phreatic morphologies and some recent, still active, vadose entrenchments. A detailed petrographic study of the host rocks has shown this cave to be formed along a set of quartz sandstone, hybrid arenites and calcarenites layers in a mostly siliciclastic sedimentary succession. XRF elemental analysis of the different rock strata guiding the speleogenesis confirms that the early stage of cave development is confined mainly in the calcium carbonate rich layers. Geochemical analyses carried out on water flowing in the cave (drips, streams) have shown that carbonate dissolution is the dominant chemical weathering process contributing to Krem Puri Cave water solutes, whereas quartz, feldspars and clays dissolution play only a secondary role in rock weathering.

The morphology of the cave points to a lithology-guided dissolution of the carbonate matrix and piping of the residual quartz sand in phreatic conditions, when the surface river network was not yet evolved and the water table was at the level of or higher than the cave. This phreatic speleogenetic phase predates the start of river network entrenchment (that started not earlier 5 Ma ago, and probably accelerated around 3.5 Ma), which deactivated the karst maze. With a low denudation rate of 0.15 mm/y, this active phreatic cave level, which currently lies beneath a more or less 120-meter overburden, was covered by at least 1-2 km of sedimentary sequence. After its decoupling from the underground drainage system, denudation slowly brought the surface close to the maze system gradually introducing infiltration vadose waters into the conduits. This caused the formation of the vadose entrenchments probably less than 100 Ka ago, and carbonate speleothem deposition in the cave.

Keywords: speleogenesis, arenites, arenization, landscape evolution, solutional weathering

1. INTRODUCTION

Caves are integral parts of an evolving landscape, being formed by dissolution (karst) and erosional processes acting in the underground (Palmer, 2009). Caves are also paleoclimatic, paleoenvironmental and geologic archives, protected from the surface morphogenetic forces, while on the topographic surface erosion and degradation often cancel most traces of the geological past. Understanding the evolution of cave systems, through the study of the hosted chemical and physical sediments, fossils, and the geomorphology of the passages, can allow to reconstruct fragments of the geological evolution of the karst area.

The morphology itself of the caves is a reflection of landscape evolution stages, especially related to changes in base level, due to regional uplift, river (or glacial) entrenchment or sea level changes, which in part can be climate-driven. A lowering base level can give rise to the formation of stacked series of cave levels, the highest being older than the lowermost, and often still active, level. Although these levels cannot be directly dated, but only their infillings (giving a minimum age of the level in which they are deposited), these chronological constraints can sometimes lead to relatively reliable models for landscape evolution (Palmer, 2007; Polyak et al., 2008; De Waele et al., 2012; Sauro et al., 2013; Columbu et al., 2018) These kinds of models are especially powerful in the reconstruction of fast-evolving karst systems, such as those in gypsum deposits (Columbu et al., 2015, 2017). In limestone areas, using cosmogenic burial dating, these reconstructions can go back into the Pliocene (Anthony and Granger, 2007), whereas using Alunite K-Ar dating the range can be extended further into the past (Polyak et al., 1998; Polyak and Provencio, 2000).

Most karst occurs in limestones and in evaporites (Ford and Williams, 2007) but also other rocks can host caves which origin is at least in part related to dissolution processes. This is the case of quartz sandstones (orthoquartzites) and metamorphic quartzites (metaquartzites), where the dissolution of minor quantities of silica triggers the formation of karst conduits that have many characteristics in common with their limestone counterparts (Wray and Sauro, 2017; Mecchia et al., 2019). The most important of these quartzarenite caves are located in South America (Venezuela, Colombia and Brazil), and their age is believed to be 20-30 Ma (Piccini and Mecchia, 2007). This age is based on a very rough calculation taking into account the quartz dissolution kinetics and solubility, the time needed for quartz grains to be released from the rock, and erosion processes to take place (Mecchia et al., 2014; 2019). The slow development of caves in this poorly soluble lithology seems to have prevented the creation of stacked cave levels, the main passage being formed along a favorable stratigraphic position (Sauro, 2014).

Until recently the Venezuelan caves in Precambrian almost pure quartz sandstones (orthoquartzites) were thought to be the only examples of large and several kilometers-long quartz sandstone cave systems in the world. The recent discovery and exploration of caves in the Shillong Plateau (Mawsynram municipality) has demonstrated the existence of likewise developed karst systems in much younger (Cretaceous) sandstones. The 25.5 km long Krem Puri (Fairy Cave) is currently the longest and most interesting of this peculiar type of caves in Meghalaya. This paper aims to address the speleogenetic mechanisms guiding the formation of a so complex maze cave and to understand the relationships between cave and landscape evolution in a region characterized by rapid uplift and fast geomorphic changes due to the high rainfall and weathering-erosional potential.

2. STUDY AREA

2.1 Geographical and climatic settings

The study area is part of the Shillong Plateau, a basement pop-up structure lying in front of the Himalayas in northeast India (Meghalaya State). This mountain range, that rises up to 1900 m asl, is the only orographic barrier separating the eastern Indian and Bangladesh Plains from the Himalayas. Because of its geographical position and the characteristics of the monsoonal climate circulation, it causes a rain shadow effect on the northern areas, whereas its southern flank is the rainiest area of the world. At Mawsynram, an average of 12 meter annual rainfall is recorded, falling mostly during the monsoon summer (in about 6 months only) (Murata et al., 2007). The highest recorded annual rainfall reached over 26 meters in 1985.

The annual rainfall recorded in 2017 and 2018 by the India Meteorological Observatory in Cherrapunji, 12 km east of Krem Puri, were 10,610 mm and 7,282 mm respectively (www.noaa.gov). In the Shillong Plateau, the meteorological year can be divided into three seasons with very unevenly distributed precipitation: pre-monsoon, monsoon and post-monsoon. In 2018, more than 97% of the total precipitation fell between the beginning of May to mid-October (monsoon season), 2% from January to the beginning of May (pre-monsoon), and less than 1%

from mid-October to the end of the year (post-monsoon). This seasonal rainfall variability is reflected in the floods of the Umiew river, that flows in the valley east of Mawsynram, that take place during June to October, with peaks mostly occurring in July to September (Wapcos Ltd., 2016).

Precipitation in the early 2018 have been less abundant than in the early 2017, with a 35% decrease in the pre-monsoon season. During the 2018 field campaign, from 18th to 28th February, the only rain occurrence in the study area was a storm in the night between 25th and 26th (21 mm recorded at the meteorological station). A very long dry period preceded this storm event, extending back to a 51 mm precipitation that fell from 10th to 12th December. That was the only rain event that occurred in the three months before the 2018 water samplings in Krem Puri. No rain fell in the cave area during the 2019 field work, that was also preceded by a very long dry period, with only 21 mm of rain (18th-19th December 2018) in the three months before the water measurements in the cave. As a result, very low water discharges were found in the cave in both 2018 and 2019 field work, and the observed water chemistry is to be taken as representative of pre-monsoon period only.

The 2018 average monthly temperatures ranged from 20°C in the summer monsoon season, to 12°C in January. In the period of 2018 measurements, the daily air temperature measured at Cherrapunji station (1313 m asl) fluctuated between a minimum of 6.4°C and a maximum of 23.2°C (average 15.3°C). In Krem Puri, where the elevations are between 1210 and 1270 m asl, the water temperatures measured at the sampling times were included in the range 13.1-21.1°C, and the air temperatures between 17.1-19.0°C. The 6th February 2019 the water temperatures measured in 6 sampling stations in the cave were in the range 12.1-19.9°C, very similar to those measured in February 2018, while the daily air temperature excursion at the Cherrapunji station was from 8.5 to 19.9°C.

2.2 Geological history

The Shillong Plateau is bordered to the south by the Bangladesh plains, to the north and west by the Brahmaputra River, and to the east by the so-called Kopati Fracture Zone (Biswas et al., 2007). This hilly plateau is bounded by two reverse and seismically active faults, the E-W striking Dauki Fault to the south and the WNW-ESE trending Oldham Fault in the north. This last was responsible for the largest historical intraplate earthquake ever registered: the June 12, 1897 Shillong earthquake was estimated at a magnitude of >8, which uplifted the northern part of the plateau by 11-16 meters (Bilham and England, 2001).

Uplift of the Shillong Plateau started as early as Middle Miocene, (14 Ma) (Biswas et al., 2007; Clark and Bilham, 2008), causing the redirection of the Brahmaputra River, and basement erosion starting around 5 Ma (Najman et al., 2016; Govin et al., 2018). Uplift accelerated between 3.5 and 2 Ma causing enhanced erosion of the basement rocks and the deposition of the fluvial Pliocene "Tipam Formation" in the Surma Basin, SE of the Shillong Plateau (Najman et al., 2016). This period coincides with the beginning of entrenchment of the main valleys and the dissection of the southern edge of the Shillong Plateau around 4.5 Ma ago (Rosenkranz et al., 2018).

2.3 Stratigraphy

The basement of the Shillong Plateau is mainly composed of metamorphic and igneous rocks of Precambrian age. In the study area, along the southern portion of the plateau, this flat basement complex is overlain by an over 1000 m thick and sub-horizontal sequence of Late Cretaceous-Tertiary sediments, dipping gently (around 5°-10°) to S or SSE.

A generalized geological map (Fig. 1) and stratigraphic description of the Mawsynram area was firstly published by Kak and Hasan (1979). However, it should be noted that their map lacks any detail of displacements and kinematics across the numerous faults intersecting the area, documented both at the surface and within the caves. Krem Puri karst system formed within the upper part of "Mahadek Sandstone Formation" (Late Cretaceous). The Formation here is approximately 450 m thick and mostly represented by grey, greenish and orange brown lithic, feldspathic and quartzose sandstones. The sandstones are fine to coarse grained and thickly bedded, with planar bedding contacts commonly with thin shale partings. Isolated ‘floating’ quartzite pebbles and cobbles are seen within the sandstone matrix in many places. This sandstone formation is underlain by a matrix-supported conglomerate known as the “Bottom Conglomerate Formation” of a thickness up to a few dozens of meters (Kak and Subrahmanyam, 2002).

The Mahadek Formation was likely deposited in a continental environment, with alluvial fans and braided rivers (Lower Mahadek Member, LMM), gradually passing upward into a shallow-to-deep marine environment (Upper Mahadek Member, UMM) (Sen, 2010). Krem Puri maze is hosted in the UMM, where the sandstones are totally devoid of cross-bedding, suggesting deep water depositional facies. Also, the ‘floating’ pebbles and cobbles indicate that turbulent debris or grain flow processes were active during transport and sedimentation (Fig. 2). Further west in the Khasi Hills region, a coastal depositional environment has also been suggested by Kak and Subrahmanyam (2002) for the upper part of UMM, reflecting paleo-geographic facies variability in the study area. Some authors also interpret a fluvial depositional environment with volcanoclastic intercalations for the LMM, where uranium mineralization has been found (Kak and Subrahmanyam, 2002; Hamilton et al., 2011).

The Krem Puri overburden comprises the upper part of the Mahadek Formation sandstones overlain by the latest Cretaceous to Lower Paleocene “Langpar and Thierra Formations”, including mixed shallow marine calcareous shales and sandstones (Jauhri and Agarwal, 2001). These are followed by a warm shallow water marine environment where the deposition of well-bedded fossiliferous “Lakadong Sandstones” and “Lakadong Limestones” occurred. These rocks sporadically outcrop at the top of some hills up to 160 m above the cave (Gogoi et al., 2009).

Stratigraphic descriptions and mineralogical studies on the rocks of the Mahadek Formation outcropping near Dauki, 30-50 km east of Mawsynram (Pathak, 1973), from Weiloi to Mawsynram (Deka, 2011), and in the West Khasi Hills (LMM, Panigrahi et al., 2013) allow to get a general idea of the lithologies in which the cave formed.

Among the detrital grains, quartz ranges between 50% to 90% and feldspar from 1% to 30% (Pathak, 1973; Deka, 2011; Panigrahi et al., 2013). Among the feldspar, K-feldspar (microcline and orthoclase) is most dominating, but sodium feldspar is also present (Deka, 2011). Alteration of feldspar to kaolinite is very common. The distribution of the detrital grain constituents in the stratigraphic sections shows that quartz becomes gradually dominant, with the impoverishment of the feldspar from the basal to the uppermost part of the Mahadek Formation (Pathak, 1973). Detrital mica (biotite, muscovite and chlorite) grains generally represent less than 1%. Other accessory minerals are iron minerals (hematite), but zircon, tourmaline, and rutile are also present (Pathak, 1973; Deka, 2011). Kaolinite, illite, montmorillonite and chlorite are the representatives of clay minerals (Pathak, 1973). The lithic fragments component, that represent the disintegrated igneous and metamorphic source rocks, ranges from 2% to 11% (Deka, 2011).

In the mainly fluvial LMM, abundance of organic matter and pyrite is observed, indicating a reducing environment (Hamilton et al., 2011). Widespread radioactivity was recorded in different layers of this member (Sen et al, 2000). High values of uranium were found in the West Khasi Hills, especially as pitchblende, derived from basement crystalline rocks like granite and granite-gneisses, and associated with low ferric/ferrous ratios, abundance of sulfates and sulfides, organic carbon and traces of selenium (Kak and Hasan, 1979; Kak and Subrahmanyam, 2002; Hamilton et al., 2011).

Regarding the cement in the Mahadek sandstones near Mawsynram, Deka (2011) found it ranging between 3% and 27%, mostly consistent of iron-oxide (hematite) that occurs either as filler of the intergranular spaces or as staining material over the grains, with fewer amounts of silica and calcite. According to Pathak (1973), in the Dauki area calcite cement is predominant in most sandstones, with percentage that reach up to 30%.

In the area of Krem Puri the Mahadek Formation dips gently by 3-4 degrees to the S or SSE. Faults and joints are mostly steeply inclined and range from hair line cracks to major fault zones likely of wrench architecture.

3. METHODS

The main study was conducted in the pre-monsoon season from 18th to 28th February 2018, and during a second mission also in the pre-monsoon period from 3rd to 7th February 2019. Samples of rocks and secondary minerals were taken with a geological hammer after documenting the sampling sites with a digital camera. Structural and geological measurements were acquired using classical survey tools (compass and clinometer). Where flowing water was present, samples were taken and in situ measurements were performed as described below.

3.1 Cave topographic survey and structural analysis

Cave surveying in Krem Puri is the result of work carried out since its discovery in 2016 by different survey teams, using the laser-based method of "Paperless Cave Surveying" (Heeb, 2009), with fixed stations. The core of this methodology is the computer software *Pocketopo* created by the Swiss engineer and speleologist Beat Heeb. The program enables the connection through Bluetooth of topographic survey instruments to a hand-held computer, where the data are stored and the polygonal segments are geo-processed in order to have a real-time topography of the cave. Manual vector graphics are enabled to produce sketches in plan, elevation and cross-sections. After the surveying campaign, data and sketches are transferred onto a computer for further processing and digital drawing.

The great advantage of a complete digital survey is that data and sketches are directly downloaded from the hand-held device to a desk or laptop computer and the random errors from manual transcriptions are therefore reduced. Also the use of a laser device reduces the errors during data acquisition compared with measurements taken using an optical compass, clinometer and tape. Provisional drawings can also be printed for immediate use and the final topography can be edited to reach the desired quality.

When surveying Krem Puri the challenge was to coordinate different teams working in different areas of the cave at the same time. Connecting different sets of data demands a strict hierarchy in survey sequences, and meticulous assignment and control of the series numbers used by the surveying teams. Connecting polygonal series needs to be unique and unambiguous, and the complex maze of Krem Puri with its hundreds of connections needed special attention by a controller.

The final topography in plan view was processed for extraction of the main directions of development using a manually controlled approach (Pisani et al., 2019; Sauro et al., 2019). The entire 25 km long system was included in the analysis with 4494 measurements of azimuth referred to magnetic north (approximated as geographic north because magnetic declination in the Meghalaya area at survey date was evaluated 0.18°W, much less than the laser meter accuracy) and length for each clearly developed cave passage > 1 m long. Measurements were grouped in classes every 5° and the calculated frequency was weighted on the overall length compared to the average length of the cave segments. This cumulative normalized frequency for each class of directions was plotted into a rose diagram using the "*Georose*" software (Yong Technology Inc., 2014), to have a quantified overview of direction trend of the whole cave system.

3.2 X-ray fluorescence (XRF) spectrometry

XRF analyses were conducted to investigate major (>1 g/100 g), minor (0.1–1.0 g/100 g) and trace (<0.1 g/ 100 g) element composition in the host rocks. An aliquot of around 3 g of each sample was milled using a mortar and pressed on rounded boric acid casts (~5 cm diameter, ~0.5 cm height), which were prepared according to the matrix correction method (Franzini et al., 1972; Leoni et al., 1982). Analyses targeted the basal thin layer of the resulting pressed powder pellets, through a sequential wavelength dispersive XRF spectrometer (Axios-Panalytical), equipped with a 4 kW Rh tube and SuperQ 3.0 software operating at the Department of Biological Geological and Environmental Sciences – University of Bologna. The settings of the instrument, as well as the standards used for calibrating the raw results, are those recently provided in Funari et al. (2015). Final results were calculated by applying the loss on ignition (LOI) estimation, measured after overnight heating at 950 °C of a further 0.5 g of each sample using a Setaram Labsys double-furnace apparatus and calcined Al_2O_3 as reference substance for the volatile content. The final accuracy was better than 5% for elements >10 ppm, and between 10% and 15% for elements <10 ppm.

3.3 X-ray diffraction (XRD)

Mineral phases in secondary sediments, wall crusts and speleothems were investigated by a Philips PW3710 X-Ray diffractometer (current: 20 mA, voltage: 40 kV, range 2θ: 5–80°, step size: 0.02° 2θ, time per step: 2 sec) at the University of Genova (Italy), which mounted a Co-anode as in De Waele et al. (2017). Minerals were ground to a fine powder in an agate mortar prior to analysis. Acquisition and processing of data were carried out using the Philips High Score software package.

3.4 Optical microscopy

Thin sections (~25 μm) were prepared at the sedimentological laboratories of Bologna University (Geology Department) from the host rock samples. They were studied under a Nikon eclipse Ci-Pol binocular petrographic microscope equipped with a 5.0-megapixel DS-Fi2-L3 digital camera operating in the above reported department.

3.5 Scanning electron microscopy (SEM)

Subsamples of sediments were first covered with a thin evaporated gold layer by sputtering, then introduced into a Vega3 Tescan scanning electron microscope and a Zeiss Supra 40 VP field emission scanning electron microscope (FESEM), operating respectively at DISTAV and DCCI departments, University of Genova (De Waele et al., 2017). The first operated at 20kV and was equipped with an EDAX-Apollo-X DPP3 energy-dispersive (EDS) X-ray spectrometer, which was applied for major elements spectrometric measurements. Manganese resolution of $K\alpha$ = 126 eV allowed the detection of chemical elements heavier than Boron (atomic number greater than 5). Acquisition and elaboration of data was performed by the TEAM Enhanced Version V4.2.2 EDS software. For FESEM images, we used accelerating voltages from 10 Å to 20kV.

3.6 Water sampling, analyses

Water temperature (T_w), electrical conductivity (EC), and acidity (pH) were measured on site in 10 stations inside the cave, by handheld field instruments (Hanna Instruments), after calibration (see Figure 6 for location of samples). Resolutions were 0.1 °C, 0.1 μS cm^{-1}, and 0.01 pH respectively. Stream discharges and dripping rates were estimated in all the stations at the sampling time. Measurements of T_w, EC, and pH in the field were also carried out in February 2019, in 6 of the same stations sampled the previous year. An alkalinity test kit (Hanna Instruments HI 3811) was used in the 2019 field work to measure water alkalinity within 12 hours of collection (expressed as phenolphthalein alkalinity and total alkalinity). Due to problems with the instrumentation it was not possible to measure alkalinity in the 2018 field campaign.

Water samples were collected in the 10 stations for the analysis of dissolved silica concentration, measured within 24 hours from sampling by using a field colorimetric test kit (Aquaquant 14410 Silicon - Merck). Double water samples were collected in each station for laboratory chemical analyses: a 250-mL bottle of untreated and unfiltered water for anion chromatography analyses, and a 100-mL bottle of 0.45 micron-filtered and 1 mL 65% HNO_3 acid-preserved water, in order to determine dissolved elements through inductively coupled plasma-mass spectrometry (ICP-MS) at Merieux Laboratories in Resana (Italy) within two weeks after sampling.

In the Merieux Laboratories, ICP-MS (method EPA 6020A) was applied for determination of dissolved concentrations of Al, Sb, As, Ba, Cd, Ca, Fe, Mg, Pb, K, Na, Zn. Recovery of the Laboratory Control Sample (LCS) resulted between 85% and 115%, as expected by the method lines. Ion Chromatography (method EPA 9056A) was used to determine chloride, fluoride, nitrate, and sulfate anions in the solution. An LCS was included in the analytical batch and the result complied the acceptance criteria (±20% of the spiked value). The molybdenum-blue spectrophotometric method was used for the determination of phosphates. The lower limits of quantitation (LOQs) are given together with the results in Tables 4 and 5. Total dissolved solids (TDS) were calculated as the total sum of dissolved ions.

To give the original composition of the water at the time of 2018 sampling, alkalinity was reconstructed via major ion charge balance, calculated as the difference between the total milliequivalents per litre of the four major cations (Na^+, K^+, Mg^{2+} and Ca^{2+}) and the sum of the anions Cl^-, SO_4^{2-} and NO_3^- expressed in the same unit. The error in a balance generally does not exceed 1% or 2% of the total ions in waters of moderate concentration (Hem, 1985). In the samples analyzed in this study the total of anions and cations is less than 5 meq L^{-1}, and a larger percentage difference is expected, maybe up to 5%. In some samples, the concentration value for one or more cation or anion was below the LOQ; in these cases, a value of half the LOQ was used for calculation. Carbonate and bicarbonate concentrations were calculated from alkalinity data by applying the “advanced speciation method” (USGS, 2012) that considers water temperature and pH. Alkalinity measurements carried out on site in February 2019 were very close to the expected values calculated for the previous year with the procedure given above.

4. RESULTS

4.1. Krem Puri description

The main entrance (E1) of Krem Puri is situated below a 60 meters high cliff, 3 km SW of Mawsynram town centre. It has two other entrances (E2 and E3), both below the cliff, respectively 900 and 1100 meters northeast from Entrance E1. E2, or ‘Balcony Entrance’, is located on a large balcony, part way down the line of a waterfall, with spectacular views out over the surrounding valleys.

Another cave belonging to the system is Krem Maduri, situated 250 m southwestward along the cliff from Entrance E1 and on the same cliff ledge and elevation (E in Figure 3). The general direction of this cave is north to south, and, although belonging to the same cave system, it fails to connect physically because of a large boulder collapse obstruction. At the closest point Krem Maduri is approximately 8-10 m higher and about 10 m horizontally from the nearest Krem Puri passage.

Most of Krem Puri passages have a typical rounded phreatic cross-section shape (Fig. 4A-C) with more or less pronounced entrenchments, many hosting small streams. The most striking feature of the cave passages is that in plan view they form a regular bi-dimensional maze, with 3 to 6 way intersections every 10 meters so. This maze develops along a single slightly inclined stratigraphic interval (Figures 4 and 8) dipping 3-4° toward the south, and almost all the passages are controlled by joint sets and faults (Figures 2 and 4).

The deepest point of the cave, ~30 meters below Entrance E1, is a narrow 13-meter-deep rift that leads to an active stream level ("Sink" in the cave map of Fig. 3), which can only be followed for around 40 m before it gets too tight. The sinking water comes from a series of hydraulically active vadose canyon passages, commonly over 10 to 20 meters deep and up to several meters wide. These canyons, along with similar abandoned ones, are the main passages in Krem Puri, generally oriented parallel to the external cliff (NE-SW & ENE-WSW). The most important of these canyons is the southernmost one, named "Stream Canyon", to which all the other streams confluence. Most of these vadose canyons have 'key-hole' shaped cross sections where it is possible to walk either on the ledges of the top section or in the narrow streamway at the bottom (Figures 4B and 4E). The ledges are often fragile and tend to break off, whereas the lower streamways are commonly blocked by fallen boulders, or too narrow to pass. Secondary canyons are typically 20 m to 30 m apart and run more or less parallel to the Stream Canyon. The smaller maze passages are mostly one meter high or less and join into the canyons at angles of around 50°. Where the canyons are deeply incised the smaller maze passages are 'perched' many meters above the canyon floor. In several parts of the cave, larger chambers have developed, mainly due to collapse around the intersections of subvertical faults, with erosion at the foot by small active streams (Fig. 4D). Total passage length in Krem Puri is around 25 km within a height range of +29 m to -26 m with respect to main entrance (E1) elevation.

Water feeds the cave either by vertical infiltration through fractures that take water from the summit surface (approximately 100 m above the cave) or by feeding into the peripheral cliff face at the known entrances and at many choked points close to the cliff. Inside the cave, very low discharges (up to few tenths of litres per minute) were observed in the pre-monsoon season in many of the streams while in the main collector a flow of several litres per minute was seen.

Many of the cave passages are fully or partially filled with fine to coarse sand with varying amounts of clay and in places calcite speleothems decorate the passages, mainly where abundant water infiltrates from above (Fig. 5A). Fossil vertebrate bones, teeth and calcareous microfossil debris are exposed in many places along the cave walls, by differential erosion and corrosion (Fig. 5B). In certain parts of the cave weakly cemented sand speleothems up to 15 cm high occur on the cave walls and floor. These sandy sculptures have been called "sandmen" by speleologists (Fig. 5C). Their origin and genesis is at present uncertain and still subject to debate. In dry parts of the cave gypsum is commonly observed as crusts and aggregates (flowers) with some fibrous growth linked to exfoliation of the cave walls and roof.

4.2 Structural and stratigraphic controls on the cave network

Krem Puri passages show a clear control provided by the intersection between a specific stratigraphic interval and fractures (Figures 2, 4 and 8). Steeply inclined or subvertical joints and faults are observed to be the main features controlling the orientations of the maze conduits within the Mahadek Sandstone and therefore the cave survey passage trends (Fig. 3) can be used as a good approximation for brittle structure orientations (Belvederi and Garberi, 1986; Calaforra et al., 1990; Stafford et al., 2008; Antonellini et al., 2019; Pisani et al., 2019). The calculated rose diagram in

Figure 7 shows two main trends, one striking ENE-WSW and the other NNE-SSW. The main one shows an average value of 65°N, while the other of 20°N.

The structural analysis with *Stereonet* software (Allmendinger et al., 2012) of the rose diagram allowed the interpretation of three different systems of conjugated fracture sets which describe the multi-deformational history and the stress field evolution in the Shillong Plateau. System A (the less relevant) is consistent with a NW-SE direction of maximum principal stress ($\sigma 1$). Systems B and C (the more relevant) are consistent respectively with NNE-SSW and a clockwise rotating NE-SW to ENE-WSW directions of maximum principal stress. These values are referable to the main compressive deformational phases recorded in the Shillong Plateau (NW-SE oldest one; NE-SW to ENE-WSW, earlier ones) (Devi and Sarma, 2010). The last NE-SW trending compressive phases are the most represented by the fracture-controlled cave passages.

The general bedding dip southward of 3 to 4° seems not to be a strong controlling element for cave passage orientation, with the main direction of entrenchment of the system slightly oblique to the bedding attitude (green shaded area in Figure 7). All the stream water converges southwestward until the bottom of the Stream Canyon branch is reached (Fig. 3). Secondary permeability provided by joints and faults, as confirmed by direct observations in the cave, plays the main guiding role for the inception and development of the phreatic conduits at the intersection with a single carbonate-cemented stratigraphic layer (Fig. 8), within a sequence where other bedding interfaces are far less conductive.

From the petrographical point of view, the study of thin sections of rock samples along the section of the cave conduits (Fig. 8) confirms the features detected in hand samples and outcrops. Indeed, all strata show medium to fine grained sandstone although with important differences in the composition of grains and cements. From the top to the bottom of the sequence reported in Figure 8, KPs1X is mostly composed of sub-rounded quartz grains together with phyllosilicate lamellas, cemented by a clay matrix (Fig. 9A). KPs1E appears as a dolomitic grainstone, with a minimal presence of sub-rounded quartz grains (Fig. 9B). Similar features are found in KPs1B. In KPs1A the major components are dolomite and quartz grains that, together with bioclasts and glauconite grains, are cemented by calcite. KPs1C is a packstone of coarse bioclasts and quartz with minimal amounts of glauconite cemented by micritic calcite. The latter can be addressed as hybrid arenites. KPs1D is mostly composed of sub-rounded quartz and feldspar grains.

The typical phreatic passage morphology in the cave reflects the local variations of petrography within a unique 2-meter-thick layer in the upper part of the Mahadek Sandstone. The main rounded (phreatic) passages of Krem Puri are represented by rock samples KPs1A, B, and C (carbonate-cemented and carbonate clasts as major components), while the narrow vadose entrenchments and fracture-guided rifts along the roof are cut through purer quartz sandstones (samples KPs1D and X). XRF results for rock samples collected in a typical cave passage are presented in Figure 8 and Table 1.

4.3 Weathering of the guiding strata and secondary minerals

Samples taken from the weathered walls of the cave along the guiding strata in Figure 8 have been observed through SEM for the detection of mineral weathering features. Quartz is typically present as pitted grains (Fig. 10A-B), with very rough and porous surfaces, probably related to secondary intense transport-controlled dissolution (Burley and Kantorowicz, 1986). Also plagioclase and K-feldspar grains show pervasive pitting often associated with kaolinite as secondary product of incongruent dissolution (Fig. 10 C-F). Calcite cement can be seen in form of fibrous prisms, often with clear dissolution morphologies (Fig. 10 G-H).

Aside of the host rock weathered strata, a total of seventeen secondary cave deposits were taken in different passages of Krem Puri and the results of SEM and XRD mineralogical analyses are reported in Table 2. Samples represent crusts of different colors on the walls, powdery deposits on the dry cave floors, and laminated sediments covering ledges and floors of passages. Sample KPm16 was a piece of strongly weathered sandstone almost turned to a sand.

Quartz is ubiquitous, often accompanied by K-feldspar, plagioclase and by minor amounts of zircon. The weathering of feldspars leads to the formation of kaolinite, which was found in two samples (KPm2 and KPm17). Carbonates are often present in minor amounts, in the form of calcite and, often, ankerite ($Ca(Fe^{++},Mg,Mn)(CO_3)_2$). The most important speleothems are composed of flowstones, stalactites and stalagmites of calcite. Minor formations (dark crusts or coralloids) are often made of phosphates (hydroxylapatite, leucophosphite, or tinsleyite). Curiously, we did not find any speleothems composed of silica. Sulfates have also been found as crusts and flowers of gypsum and traces of barite, probably related to the oxidation of pyrite flecks in the sandstones. Phosphates are present in the form of hydroxylapatite and, sometimes, of leucophosphite and tinsleyite.

4.4 Water analysis

The results for physical parameters and major ion concentrations, measured or calculated, are reported in Table 3. Concentrations of the analyzed metals are shown in Table 4. For sample location see Figure 6.

The analyses of major ions represented by bar lengths in milliequivalents per litre are shown on the cave map of Figure 6, while for heavy metals in Figure 11 the bar lengths represent milligram per litre. Major ions concentration is also represented in a trilinear diagram (Fig. 12).

The geochemical code PHREEQC v.3.0 (Parkhurst and Appelo, 2013) and its integrated databases were used to estimate saturation indices (SI) of ground water with respect to minerals likely to be present within the aquifer. These are defined as $SI = log\left(\frac{IAP}{K_{sp}}\right)$, where IAP is the ion activity product, and K_{sp} is the solubility product constant. Many of the features of the chemical evolution of ground water can be explained in terms of mineral dissolution and precipitation reactions. SI indicates if a solution is in equilibrium with a solid phase or if under-saturated and super-saturated in relation to a solid phase, and helps to assess the evolution stage of a groundwater sample and identify the controlling geochemical reactions.

The mineral assemblage of rocks in the study area can be compared with the chemical composition of waters to characterize the processes of water-rock interaction in the area. The minerals chosen as controlling phases to represent the study area lithology are the following: quartz and amorphous silica, K-feldspar, albite, kaolinite, illite, Ca-montmorillonite, chlorite, K-mica, calcite, dolomite, gypsum, hematite, goethite, and amorphous ferric hydroxide. Results are presented in Table 5 for the silicates, and in Table 6 for carbonates, sulfates, and oxyhydroxides. In a number of samples, aluminium and iron are at concentrations below their limits of quantification of 0.02 and 0.01 mg/L respectively. Chloride, potassium, sodium, and magnesium are also below their detection limit of 0.4 mg/L in a few samples. In all these cases, to determine the possible range of saturation indices of minerals, two simulations were run with PHREEQC, the first entering their LOQ values as the concentrations (that give the maximum possible saturation indices), and the second entering values equal to 0.1 LOQ (assumed as minimum possible SIs). It should be noted that a saturation index does not necessarily demonstrate whether mineral dissolution or precipitation actually happens.

Geochemical evolution models and mass transfer due to chemical reactions in groundwaters are generally characterized along hydrologic flow paths. This approach can reveal systematic variations in the concentration of major elements, and the Total Dissolved Solids (TDS) are expected to gradually increase from recharge areas along the flow path due to mineral dissolution.

Considering vadose water flow in the unsaturated zone of an aquifer, expected to be homogenous in terms of mineralogical composition, TDS increases essentially with the time length of water-rock interaction in fractures, which in turn depends on the travelled distance and the mechanical characteristic (mainly width) of the fracture in which the water flows (Mecchia et al., 2019). The vertical distance between the ground surface (the recharge area on the plateau) and the cave level in which drips and seepage water samples were collected ranges up to approximately 100 m. Our geochemical evolution modelling approach thus assumes that higher TDS represents more evolved water moving along a single virtual geochemical evolution flow path, in which the results of all the water samples collected in the cave are included. A conceptual graphic illustration of the real flow paths hypothesized for the water in Krem Puri is shown in Figure 13, where the length of each flow path justifies the TDS content found in the samples.

Following this approach, the behavior along the geochemical evolution path is represented in Figure 14, where the relationships between major cations and TDS are shown on the left, while on the right the changes in the saturation indices and their linear trends for the most important minerals, assumed to be present in the aquifer, are represented. Plots Ca^{2+}+Mg^{2+} vs. HCO_3^- (Fig. 15A), and Ca^{2+} vs. Mg^{2+} have been developed (Fig. 15B) to improve the understanding of calcite and dolomite dissolution in the upper part of the Mahadek Formation vadose aquifer.

The concentrations of carbonate and bicarbonate ions in water samples were calculated from sample pH (maximum value 8,1) and alkalinity (Table 4) according to theoretical relations based on the chemistry of water and carbonic acid, according to the advanced speciation method (USGS, 2012). The results show very low carbonate concentrations, below 0.7 mg/L in all the samples.

Inverse modelling (PHREEQC, Parkhurst and Appelo, 2013) was used to reconstruct the geochemical evolution of waters through the vadose zone of the Krem Puri aquifer from their chemical compositions, and investigate the control of lithology on the water chemistry. Inverse modelling attempts to account for the chemical changes that occur as water evolves along a geochemical evolution path, assuming two water analyses which represent starting and ending water compositions along the evolution path. The water sample analyses with the lowest and the highest TDS values were chosen to represent initial and final solutions in the system. Since the stoichiometry of trace elements is not well known, and trace elements can be affected by sorption to mineral surfaces, our inverse modelling simulation included the major elements but not the trace elements. The other model input parameters consist of field-measured pH, temperature, and dissolved SiO_2. Application of inverse modelling requires knowledge of specific mineral compositions. The phases in the input files represent minerals which are susceptible to weathering as shown in SEM images, and minerals detected in XRD analysis likely to precipitate as alteration products. We chose to include K-feldpsar (Kfs), albite (Ab), kaolinite (Kln), K-mica (Ms), illite (Ill), calcite (Cal), dolomite (Dol), gypsum (Gy), amorphous silica and CO_2. CO_2 was included as a gaseous phase since it participates in most weathering reactions. The weathering of quartz is so slow that it is impossible to describe it using equilibrium reactions, moreover, quartz resulted close to equilibrium, suggesting that it probably participates minimally in chemical reactions. The inverse model was constrained so that primary mineral phases including calcite, dolomite, albite, and CO_2 were set to dissolve until they reached saturation. A number of models were obtained with each modelling exercise run with PHREEQC. These models reconstruct all possible combinations of dissolution and/or precipitation reactions that explain the chemical changes observed between the two solutions and the mineral phases. A selection of the results for the best modelling runs is presented in Figure 16, and they can be used to examine the influence of lithology on the dissolution and precipitation of mineral phases, showing the moles transferred from each phase, dissolved or precipitated.

5. DISCUSSION

5.1 Stratigraphic and structural control on karst porosity in the Mahadek Sandstones

In contrast to the Mahadek Sandstone as a whole within Krem Puri an interval up to 2 m thick is carbonate-cemented rather than being iron-oxide or silica-cemented (see also Figure 8 and Table 1). Also in the same interval, there are both siliceous and carbonate interbeds with coarser clasts making up a calcarenite or calcirudite interval. One unusual widespread coarser bed, shown in Figure 2 comprises a mixture of quartzite and sandstone pebbles and cobbles and transported limestone clasts. This bed is also rich in disarticulated reptile bones, and reptile and shark teeth (Fig. 5B). The limestone clasts show evidence for being soft at the time of deposition and then compacted together soon after burial. This narrow calcareous package of strata (KPs1 E-B-C, Fig. 8) has likely acted as an important phreatic inception horizon (Lowe, 1992; Filipponi et al., 2009) guiding the primary proto-conduits forming the cave maze. This phreatic phase was followed by downward vadose canyon incision into the underlying purer quartz-rich strata, and upwards roof collapse. Because the areal extension of the calcium carbonate-rich bed is widespread (>1 km^2) it likely formed as a deep marine fan deposit in a slope or foot of slope setting.

The arrangement and distribution of phreatic proto-conduits shows a combination of dissolution controlled by stratigraphic variations (probably guided by carbonate matrix dissolution reflecting porosity/petrographic variations) and sub-vertical fractures. Conversely, the development and enlargement of passages in vadose conditions, together with erosional and collapse processes, are clearly focused along secondary permeability provided by tectonic structures (joints and faults).

The development of an almost horizontal cave maze in the more soluble carbonate-cemented sandstone has been most probably related to the processes of fracture feeding aquifers described by Palmer (1975). When below the water table, the fracture network was supplied uniformly to all major fractures, each one experiencing the same weathering/dissolution potential. However, the weathering/dissolution potential differs along the vertical fractures due to changes in the mineral composition of the sandstone sequence. Because of that, the proto-conduits formed only at the intersection between the fracture network and the carbonate-cemented sandstone layer, since this layer intrinsically displayed the highest potential for dissolution due to the higher presence of carbonate cement.

5.2 Chemical weathering processes in carbonate-rich sandstones

The waters analyzed are related to the present day vadose pathways above and inside the cave system during the pre-monson period. Even though they do not represent the phreatic waters responsible for the primary speleogenesis of the cave network, their characteristics provide important clues on the weathering processes that are acting in these sandstones along the fracture network and cave passages.

The ionic compositions of the sampled waters are classified into the calcium-bicarbonate type (Fig. 11). The water in pool of sample KPw7 represents an exception, being classified as mixed magnesium-calcium-bicarbonate type. This water sample has been excluded from the set of data analyzed in the following sections, because contamination before sampling is suspected. All the water samples show low mineralization, from 42 mg/L in dripping water of station KPw8, to 144 mg/L in dripping water of station KPw6.

Calcite and dolomite

The plot of (Ca^{2+}+Mg^{2+}) vs. HCO_3^- (Fig. 15A) indicates a strong positive linear correlation that follows calcite and dolomite dissolution trends:

$$CaCO_3 + H_2O + CO_2 \leftrightarrow Ca^{2+} + 2HCO_3^- \quad \text{(calcite)}$$

$$CaMg(CO_3)_2 + 2H_2O + 2CO_2 \leftrightarrow Ca^{2+} + Mg^{2+} + 4HCO_3^- \quad \text{(dolomite)}$$

where CO_2 derives mainly from biological processes in the soil zone (e.g. organic matter decomposition, root respiration, etc). Calcite and dolomite saturation indices increase along the evolution paths, indicating dissolution (Fig. 14 right). The molar proportions of [Ca^{2+}+Mg^{2+}] to 2[HCO_3^-] in Figure 10A is linear, with a slope of 1, meaning that calcite and dolomite dissolve according to the given equations. Figure 14B shows the relationships between Ca^{2+} and Mg^{2+}, where the average ratio [Mg^{2+}]/[Ca^{2+}] is 29%. This finding could reflect a low content of dolomite or high magnesium limestone in the Mahadek Formation, or the relatively short residence time of the water, since dolomite dissolution is much slower than that of calcite.

Inverse modelling (Fig. 16) attempts a quantification of the dissolution of 0.623 mmol of calcite per kg of water vs. 0.063 mmol of dolomite per kg of water along the geochemical evolution path chosen to represent the vadose flow by percolation through the unsaturated aquifer of the Mahadek Formation down to the Krem Puri cave level.

Figure 14 right shows that calcite saturation is reached in water sample KPw6, where in fact calcite precipitates forming a flowstone and showing that the water can remain in contact with calcite cement long enough to dissolve calcite until equilibrium. It should be noted that when water reaches saturation with respect to calcite, it is still undersaturated with respect to dolomite, or dolomitic limestone.

Feldspars and their weathering products

Since the weathering of silicate minerals is a slow process, the changes in water chemistry are also expected to be slow and less extensive than in carbonates. In the area of study, K-feldspar is generally the dominating feldspar, but albite is also present (Deka, 2011). The effect of their weathering on the water chemistry is primarily the addition of K^+ and Na^+ cations, and silica. The alteration product is mostly kaolinite, and probably also illite and montmorillonite (Blum, 1994). All the waters contained in the sandstone cave are undersaturated with respect to albite and also to K-feldspar (Table 5 and Fig. 14 right).

The plot of Na^+ vs. TDS (Fig. 14 left) shows an increase of Na^+ along the evolution path. Na^+ should mainly derive from weathering of albite. This result is also reflected in the increase of the albite saturation index along the evolution path (Fig. 9 left). Inverse modelling result shows (Fig. 15) modest albite dissolution (average 0.033 mmol kg^{-1} H_2O).

The plot of K^+ vs. TDS (Fig. 14 left) does not show clear changes along the evolution path in the cave. However, a general increase of the saturation index is calculated by PHREEQC for K-feldspar along the evolution path (Fig. 13 right) indicating probable dissolution. The inverse modelling result (Fig. 15) shows very modest K-feldspar dissolution in Krem Puri (average 0.012 mmol kg^{-1} H_2O). Figure 17 shows the silicate stability diagram, calculated with thermodynamic data from Robie et al. (1979) and containing stability fields for the K-feldspar microcline and its possible weathering products gibbsite, kaolinite and muscovite, expressed as a function of log ($[K^+]/[H^+]$) and log ($[H_4SiO_4]$). According to that, kaolinite is the stable phase in all sampled waters (as in most surface or near-surface continental waters), except KPw10. This sample was taken a few meters from Balcony Entrance E2, fed by the waterfall on the external cliff. In stream water inside the cave, kaolinite is supersaturated (Tab. 7, Fig. 13 right), but the saturation indices along the evolution paths are rather scattered. Inverse modelling does not give a clear result, although the average result (-0.003 mmol kg^{-1} water) suggests a very slight conversion of feldspar into kaolinite.

Quartz and amorphous silica

The plot of SiO_2 vs. TDS (Fig. 13 left) presents a slight increase along the evolution path. According to the calculations run by PHREEQC, waters in the area of study generally result around equilibrium with respect to quartz (Table 7, Fig. 13). However, PHREEQC for quartz uses a solubility product of $K=10^{-3.98}$ at 25°C, while Rimstidt (1997) suggested a higher value, $K=10^{-3.72}$. If this value were used to calculate the saturation indices, only a few samples would have resulted supersaturated with respect to quartz. SEM evidences of pitted quartz grains due to solutional weathering suggest that the second value of K is probably more appropriate in natural settings. Nonetheless, although quartz is by far the dominant component of the Mahadek Sandstones, only a limited dissolution is expected, because other minerals with much higher dissolution rates are present in significant amounts in the rocks of the study area.

5.3 Arenization and piping in carbonate-rich sandstones

While saturation indices in cave waters indicate that most of the sandstone mineral elements are affected by dissolution, it is clear that the different solution rates of mineral elements drive a progressive increase in porosity and a decrease in cohesion of the sandstones. The dissolution of the calcium/magnesium carbonate cement tends to release the other mineral grains and to turn the rock into a fragile material easy to erode. On the other hand, the incongruent dissolution of feldspars produces clay minerals that can fill the porosity and keep the weathered rock partially bounded. While arenization in pure quartz sandstone produces exclusively high amounts of quartz sands (Sauro, 2014; Mecchia et al., 2019) that are afterwards removed by piping mechanisms, in the case of Krem Puri sandstones the cave passages show the presence of important residual sediments constituted by a mixture of quartz and feldspar grains together with clay minerals (Table 2). It is also important to notice that the presence of gypsum crusts on the cave walls probably indicates that sulfide (pyrite) oxidation occurs at places, leading to replacement of calcite by gypsum with the release of carbon dioxide, according to the typical sulfuric acid reaction (Tisato et al., 2012; D'Angeli et al., 2019). This additional weathering process might have taken place in the early phases of speleogenesis, but could still be active today.

Despite the numerous active solutional weathering processes, in comparison with other studied sandstone and orthoquartzite caves, walls do not show pervasive arenization in Krem Puri. This could be due to the fact that the weathering front cannot penetrate significantly inside the rock because of the limited porosity controlled by the formation of secondary mineral crusts and by the presence of a clay matrix in addition to the carbonate cement.

It is most probable that in carbonate-rich sandstone solutional weathering (arenization) and piping/erosion progress at the same pace, but the phreatic network must be characterized by sufficient hydraulic gradient evacuating the insoluble material and enlarging the cave passages. It is probable that Krem Puri represents only a minimal relict of an ancient very large phreatic network, fed by diffused infiltration through the fracture system.

5.4 Cave and landscape evolution

The Shillong Plateau became a topographic high around 5 million years (Ma) ago, causing the redirection of the Brahmaputra River which now flows along the northern and western edge of the plateau before running south and merging into the Ganges (Govin et al., 2018). The incision of a fluvial network started at that time, but denudation rates on the plateaus were not able to match up with the higher uplift rate (Fig. 18 A), especially during the period of accelerated uplift of the Pliocene (3.5-2.0 Ma) (Najman et al., 2016). According to this rapid uplift, the topographic high of Shillong became a rain barrier and denudation rates gradually increased, eventually balancing the mountain uplift rates. An equilibrium state was established approximately 1 Ma ago, when the

Shillong Plateau reached its actual 1,600 m a.s.l. elevation (Rosenkranz et al., 2018). ^{10}Be-derived catchment-wide erosion rates on upstream (plateau) samples taken in our study area (SWC samples in Rosenkranz et al., 2018) vary between 0.14-0.16 mm/y.

Krem Puri nowadays lies at 1,230 m a.s.l., around 120 meters below the plateau surface (Fig. 18 B). Geomorphological evidences point to a first stage of speleogenesis as phreatic system (Fig. 4), hence occurring below or at the same level of an ancient water table. The phreatic maze was deactivated when the local river network entrenched, lowering the water table (Fig. 18 A). This could have happened only when the Shillong plateau became a topographic high, around 5 Ma, and probably much later when the topographic gradient became high enough.

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The local base level (River Umngi) below Krem Puri cave is now located at 500 m a.s.l., showing river entrenchment to have occurred over 700 meters since the cave was deactivated (Fig. 18 B). Using the average erosion rate measured in these deep river beds of 0.05-0.1 mm/y (Rosenkranz et al., 2018; Strong et al., 2019), 7-14 Ma would have been necessary to carve this canyon. This is incompatible with the presence of the phreatic maze system that cannot be older than 5 Ma. If entrenchment started 5 Ma ago, erosion rate in the river would have been 0.14 mm/y, similar to the values obtained by Rosenkranz et al. (2018). Using the faster erosion rates (0.4-0.53 mm/y) reported in Biswas et al. (2007) the river entrenchment, and thus cave maze abandonment, would have happened in 1.5 Ma ago. The age of the cave probably lies somewhere in between, and might coincide with the onset of the higher uplift rates since 3.5 Ma (Najman et al., 2016).

Using an average erosion rate on the plateau of 0.15 mm/y, a total of 1-2 km of overlying rocks were lost by surface erosion during this time span (5 Ma), so the phreatic maze system was originally at least 1 kilometer below the surface. Using the faster uplift (and thus erosion) rates of Biswas et al. (2007) the depth of the cave system would have been triple (3-4 km).

Since the deactivation of the phreatic phase the plateau and the cave within were uplifted for ca. 700 meters, surface canyons formed and deepened, and scarps retreated. The phreatic tubes were later excavated by vadose 15 m deep canyons that are still active today. However, the origin of this

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vadose entrenchment cannot be straightforwardly associated to the deactivation of the phreatic phase considering that: i) if vadose excavations started at that time, lasting for Ma-timescale, they would have been much more deeper than 15 m. Indeed, using both average and maximum erosion rates of 0.15 mm/y and 0.50 mm/y, these canyons would have formed in 100,000-30,000 years, a period too brief to be compatible with their carving immediately after base level lowering; and ii) they are currently active, so the morphogenetic process associated to hydrological circulation must be of young origins (i.e. not fossils). Additionally, lying deep below the surface, once this maze system was cut off from the base-level river, the cave passages appear to have remained dry for most of the time, with probably no infiltration water reaching these deep voids. The overlying bedded succession of fine and coarse thickly bedded sandstones, calcareous shales and limestones did not allow infiltration waters to penetrate deep enough and reach the phreatic maze cave. Surface waters may have been drained through limestone caves above, or simply created surface runoff, as happens today on the sandstone plateaus. With the above lying surface getting closer, infiltration waters started to enter the maze system through fractures deep enough to reach the cave, forming underground streams that started entrenching the original maze along the main open fractures in vadose conditions. For these reasons, we believe this process to have occurred rather recently and possibly over the last 100,000 years.. The moderate size of the calcite flowstones decorating few parts of the cave is in agreement with this recent age estimate.

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6. CONCLUSIONS

The discovery of the 25-km long Krem Puri has made it the longest sandstone cave in the world, exceeding Imawari Yeuta Cave in Venezuela by over 6 km (Wray and Sauro, 2017). The cave has developed along a specific stratigraphic horizon, similar to what occurs in orthoquartzite caves described in other continents (Wray and Sauro, 2017). These beds can be defined as inception horizons (Lowe, 1992), in which solutional weathering was faster, thus enhancing the development of proto-conduits (karst porosity) connecting the main underground drainage network. The main characteristic of Krem Puri is the distribution of small phreatic passages in a dense geometric and fracture-controlled maze, later cut by 15-m deep vadose canyons.

A petrographic survey of this guiding stratigraphic horizon has shown the presence of a 1 to 2 m thick carbonate cemented sandstone, hosted within a prevalently quartzose sandstone succession. Karst porosity is controlled by the secondary permeability pathways provided by the local fracture network. The interconnectivity of conduits formed a geometric dense maze imposed on a conjugated joint system (ENE-WSW and NNE-SSW) consistent with direction of principal stress oriented around a mean value of 43°N ± 2° and related to the main compressional phases recorded in the Shillong basin (Devi and Sarma, 2010).

The water geochemistry shows a Ca-CO_3 character indicating dissolution of calcite to be the major source of their dissolved load. Minor contributions from K-feldspar, kaolinite and quartz dissolution are however detected, but are largely subordinate respect to the dissolution of calcite and, to a minor degree, dolomite.

The phreatic maze of Krem Puri formed in a period when the water table was higher than or close to the cave level. Base level is actually 700 meters lower than the cave, and using the erosion rates of 0.14 mm/y (Rosenkranz et al., 2018) and 0.50 mm/y (Biswas et al., 2007) the deactivation of the cave can be estimated between 5 and 3.5 Ma. Whereas at 5 Ma the Shillong Plateau became a topographic high, relief evolution and river entrenchment was probably more important from 3.5 Ma onward (Najman et al., 2016). Using the lower erosion rates of 0.15 mm/y, this means that the phreatic drainage network, of which Krem Puri is a part, probably developed as deep as 1-2 km below the surface. Recent infiltration of surface waters probably started rather recently (less than 100 ka), since the vadose canyons that cut the original phreatic maze are not very deep (up to 15 meters) and calcite speleothems do not have significant sizes.

This study shows that important phreatic cave mazes (karstic macro porosities) can develop for tens of kilometers at the intersection between fracture networks and carbonate cemented layers bounded by quartz sandstones, with implications also for the interpretation of macro porosities in confined oil reservoirs in sandstone stratigraphic sequences (Bjørlykke et al., 2010).

Acknowledgements

The cave survey was realized thanks to the effort of a wide team of speleologists under the "Abode the Cloud Project" organization.

References

Allmendinger, R. W., Cardozo, N., Fisher, D., 2012. Structural geology algorithms: Vectors and tensors in structural geology. Cambridge University Press, UK.

Anthony, D. M., Granger, D. E., 2007. A new chronology for the age of Appalachian erosional surfaces determined by cosmogenic nuclides in cave sediments. Earth Surface Processes and Landforms 32(6), 874-887.

Antonellini, M., Nannoni, A., Vigna, B., De Waele, J., 2019. Structural control on karst water circulation and speleogenesis in a lithological contact zone: The Bossea cave system (Western Alps, Italy). Geomorphology 345, article 106832.

Belvederi, G., Garberi, M.L., 1986. Preliminar observation on the relationships between tectonic structure and genetical development of the gypsum karst cavities (Farneto, Bologna - Italy). Le Grotte d'Italia 4(12), 33-37.

Bilham, R., England, P., 2001. Plateau 'pop-up' in the great 1897 Assam earthquake. Nature 410 (6830), 806-809.

Biswas, S., Coutand, I., Grujic, D., Hager, C., Stockli, D., Grasemann, B., 2007. Exhumation and uplift of the Shillong Plateau and its influence on the Eastern Himalayas: new constraints from apatite and zircon (U–Th–(Sm))/He and apatite fission track analysis. Tectonics 26, TC002125, 22 p.

Blum, A.E., 1994. Feldspars in weathering. In: Parsons, I. (Ed.), Feldspars and Their Reactions. NATO ASI Series. Springer, Netherlands, pp. 595–630.

Bjørlykke, K. and Jahren, J., 2010. Sandstones and sandstone reservoirs. In Petroleum Geoscience (pp. 113-140). Springer, Berlin, Heidelberg.

Burley, S.D., Kantorowicz, J.D., 1986. Thin section and S.E.M. textural criteria for the recognition of cement-dissolution porosity in sandstones. Sedimentology 33(4), 587-604.

Calaforra, J. M., Pulido-Bosch, A., Sanchez-Martos, F., Montero-Lopez, A., 1990. Geomorfologia y estructura del sector de la Cueva del Agua, karst en yesos de Sorbas. Actas del V Congreso Espanol de Espeleologia, Camargo-Santander 1-4 noviembre 1990, p. 447-453.

Clark, M. K., Bilham, R., 2008. Miocene rise of the Shillong Plateau and the beginning of the end for the Eastern Himalaya. Earth and Planetary Science Letters 269(3-4), 337-351.

Columbu, A., De Waele, J., Forti, P., Montagna, P., Picotti, V., Pons-Branchu, E., Hellstrom, J., Bajo, P., Drysdale, R., 2015. Gypsum caves as indicators of climate-driven river incision and aggradation in a rapidly uplifting region. Geology 43(6), 539-542.

Columbu, A., Chiarini, V., De Waele, J., Drysdale, R., Woodhead, J., Hellstrom, J., Forti, P., 2017. Late quaternary speleogenesis and landscape evolution in the northern Apennine evaporite areas. Earth Surface Processes and Landforms 42(10), 1447-1459.

Columbu, A., Sauro, F., Lundberg, J., Drysdale, R., De Waele, J., 2018. Palaeoenvironmental changes recorded by speleothems of the southern Alps (Piani Eterni, Belluno, Italy) during four interglacial to glacial climate transitions. Quaternary Science Reviews 197, 319-335.

D'Angeli, I. M., Nagostinis, M., Carbone, C., Bernasconi, S. M., Polyak, V. J., Peters, L., McIntosh, W., De Waele, J., 2019. Sulfuric acid speleogenesis in the Majella Massif (Abruzzo, Central Apennines, Italy). Geomorphology 333, 167-179.

Decker, D. D., Polyak, V. J., Asmerom, Y., Lachniet, M. S., 2018. U–Pb dating of cave spar: A new shallow crust landscape evolution tool. Tectonics 37(1), 208-223.

Deka D.J., 2011. Characterization of the cretaceous tertiary K-T transition boundary in the Southern part of the Shillong Plateau along Weiloi-Mawsynram section, India. Unpublished Ph.D. thesis of Gauhati University, http://hdl.handle.net/10603/114298.

Devi, N.R., Sarma, K., 2010. Strain analysis and stratigraphic status of Nongkhya, Sumer and Mawmaram conglomerates of Shillong basin, Meghalaya, India. Journal of Earth System Science 119, 161-174.

De Waele, J., Ferrarese, F., Granger, D., Sauro, F., 2012. Landscape evolution in the Tacchi area (central-east Sardinia, Italy) based on karst and fluvial morphology and age of cave sediments. Geografia Fisica e Dinamica Quaternaria, 35 119-127.

De Waele, J., Carbone, C., Sanna, L., Vattano, M., Galli, E., Sauro, F. and Forti, P., 2017. Secondary minerals from salt caves in the Atacama Desert (Chile): a hyperarid and hypersaline environment with potential analogies to the Martian subsurface. International Journal of Speleology, 46(1): 51.

Mecchia, M., Sauro, F., Piccini, L., Columbu, A., De Waele, J., 2019. A hybrid model to evaluate subsurface chemical weathering and fracture karstification in quartz sandstone. Journal of Hydrology 572, 745-760.

Murata, F., Hayashi, T., Matsumoto, J., Asada, H., 2007. Rainfall on the Meghalaya plateau in northeastern India – one of the rainiest places in the world. Natural Hazards 42, 391-399.

Najman, Y., Bracciali, L., Parrish, R. R., Chisty, E., Copley, A., 2016. Evolving strain partitioning in the Eastern Himalaya: The growth of the Shillong Plateau. Earth and Planetary Science Letters, 433, 1-9.

Palmer, A. N., 1975. The origin of maze caves. National Speleological Society Bulletin 37, 56-76.

Panigrahi B., Chopra R., Mukherjee S., Singh R.V., Joshi G.B., 2013. Lithofacies controlled uranium mineralisation in Lower Mahadek sandstones of Mahadek Formation, Wahkut area, West Khasi Hills district, Meghalaya. Jour. Indian Association of Sedimentologists 32(1-2), 41-49.

Unknown
Field Code Changed

Parkhurst, D. L., Appelo, C. A. J., 2013. Description of input and examples for PHREEQC version 3: a computer program for speciation, batch-reaction, one-dimensional transport, and inverse geochemical calculations. US Geological Survey numbered series, 6-A43.

Pathak, C., 1973. A study on the Cretaceous formation in and around Dawki, Meghalaya. Unpublished Ph.D. thesis of Gauhati University, http://hdl.handle.net/10603/69967.

Piccini, L., Mecchia, M., 2009. Solution weathering rate and origin of karst landforms and caves in the quartzite of Auyan-tepui (Gran Sabana, Venezuela). Geomorphology 106(1), 15-25.

Pisani, L., Antonellini, M., De Waele, J., 2019. Structural control on epigenic gypsum caves: evidences from Messinian evaporites (Northern Apennines, Italy), Geomorphology 332, 170-186.

Polyak, V. J., Provencio, P. P., 2000. Summary of the timing of sulfuric acid speleogenesis for the Guadalupe caves based on ages of alunite. Journal of Cave and Karst Studies 62(2), 72-74.

Polyak, V. J., McIntosh, W. C., Güven, N., Provencio, P., 1998. Age and origin of Carlsbad Cavern and related caves from $^{40}Ar/^{39}Ar$ of alunite. Science 279(5358), 1919-1922.

Polyak, V., Hill, C. and Asmerom, Y., 2008. Age and evolution of the Grand Canyon revealed by U-Pb dating of water table-type speleothems. Science, 319(5868): 1377-1380.

Rimstidt, J. D., 1997. Quartz solubility at low temperatures. Geochimica et Cosmochimica Acta 61(13), 2553-2558.

Robie, R. A., Hemingway, B. S., Fisher, J. R., 1979. Thermodynamics of Minerals and related Substances. Geological Survey Bulletin, 1452.

Rosenkranz, R., Schildgen, T., Wittmann, H., Spiegel, C., 2018. Coupling erosion and topographic development in the rainiest place on Earth: Reconstructing the Shillong Plateau uplift history with in-situ cosmogenic ^{10}Be. Earth and Planetary Science Letters 483, 39-51.

Sauro, F., 2014. Structural and lithological guidance on speleogenesis in quartz-sandstone: Evidence of the arenisation process. Geomorphology 226, 106-123.

Sauro, F., Zampieri, D., Filipponi, M., 2013. Development of a deep karst system within a transpressional structure of the Dolomites in north-east Italy. Geomorphology 184, 51-63.

Unknown
Field Code Changed

Sauro, F., Mecchia, M., Piccini, L., De Waele, J., Carbone, C., Columbu, A., Pisani L., Vergara, F., 2019. Genesis of giant sinkholes and caves in the quartz sandstone of Sarisariñama tepui, Venezuela. Geomorphology 342, 223–238.

Sen D.B., Swarnkar B.M., Singh J., Dwivedy K.K., 2000. Characterisation of Ore-grade Uranium Mineralisation in the Cretaceous Lower Mahadek Sediments, Meghalaya, India. Memoir Geological Society of India, No. 46, pp 383-396.

Sen S., 2010. A contribution towards stratigraphy and palaeontology of the Cretaceous sedimentaries in Khasi and Jaintia hills Meghalaya India. Unpublished Ph.D. thesis of Gauhati University, http://hdl.handle.net/10603/114342.

Unknown
Field Code Changed

Stafford, K.W., Nance, R., Rosales-Lagarde, L., Boston, P.J., 2008. Epigene and hypogene gypsum karst manifestations of the Castile Formation: Eddy County, New Mexico and Coulberson County, Texas, USA. International Journal of Speleology 37(2), 83-89.

Strong, C. M., Attal, M., Mudd, S. M., Sinclair, H. D., 2019. Lithological control on the geomorphic evolution of the Shillong Plateau in Northeast India. Geomorphology 330, 133-150.

Tisato, N., Sauro, F., Bernasconi, S. M., Bruijn, R. H., De Waele, J., 2012. Hypogenic contribution to speleogenesis in a predominant epigenic karst system: a case study from the Venetian Alps, Italy. Geomorphology 151, 156-163.

U.S. Geological Survey, 2012. Techniques of Water-Resources Investigations (TWRI). Book 9. Section A. National Field Manual for the Collection of Water-Quality Data.

Wapcos Ltd., 2016. Comprehensive environmental impact assessment study for Mawphu hydroelectric project (85 MW), Stage II, (I).

Woodhead, J., Pickering, R., 2012. Beyond 500 ka: Progress and prospects in the U/Pb chronology of speleothems, and their application to studies in palaeoclimate, human evolution, biodiversity and tectonics. Chemical Geology 322, 290-299.

Wray, R. A., Sauro, F., 2017. An updated global review of solutional weathering processes and forms in quartz sandstones and quartzites. Earth-Science Reviews 171, 520-557.

Rick Allmendiger's stuff, Stereonet v.9, http://www.geo.cornell.edu/geology/faculty/RWA/programs/stereonet.html.

Yong Technology Inc., 2014, GeoRose, http://www.yongtechnlogy.com/download1/georose/.

Figure captions

Figure 1. General location of the study area and geological map (modified after Kak and Hasan, 1979; Gogoi et al., 2009; Deka, 2011).

Figure 2. Stratigraphic features observed in the cave: A. The typical "8" shaped phreatic conduit with sub-horizontal bedding planes (yellow lines) and sub-vertical guiding-fractures (Photo Daniela Barbieri); B. These beds are often composed of chaotic sandstones with high amounts of carbonate cement (Photo Mark Tringham); C. Some interlayers include decimeter-sized quartz cobbles in a sandy matrix (Photo Mark Tringham).

Figure 3. Plan view of Krem Puri cave. Orange-brown arrows show the sandstone cliff edge, orange arrows (E1-E3 and E) the cave entrances, blue and yellow lines are the main and secondary drainage routes. All other cave passages (grey) are not hydrologically active anymore. The lower schematic cross-section is WSW-ENE directed. The vertical purple arrows indicate the areas where water sinks.

Figure 4. Cave morphology (dashed white line = fractures and bedding; full white line = phreatic maze passage outlines; full yellow line = vadose canyon passage outlines): A. The phreatic rounded tunnels ending at the top of a deep vadose canyon passage (Photo Chris Howes); B. A main rounded phreatic passage cut by a later-stage vadose canyon (note guiding-fractures on the roof) (Photo Mark Burkey); C. The rounded phreatic passages can be only a few meters apart, but generally are regularly spaced and almost parallel (Photo Toby Hamnett); D. The largest chambers in Krem Puri are mostly formed by breakdown with subvertical joints or faults on the roof and walls and large blocks on the ground (Photo Mark Burkey); E. Upper rounded phreatic passage above, and vadose downcutting canyon below (Photo Mark Tringham).

Figure 5. Deposits and fossils: A. One of the rare calcite speleothems in Krem Puri, a more than 4-meter-high flowstone (Photo Mark Tringham); B. Cretaceous aged fossil teeth and bones of unidentified marine vertebrates in the passage wall (Photo Marcel Dikstra); C. The curious sedimentary deposits named "sandmen", likely related to plastic deformation or turbulent water flow (Photo Marcel Dikstra).

Figure 6. Analyses of water samples represented by bar lengths in milliequivalents per litre. Dissolved silica concentration (SiO_2), electrical conductivity (EC), pH, and estimated water discharge (Q) are indicated near the bars.

Figure 7. Rose diagram of Krem Puri passage trends based on cave survey processing and statistical analysis.

Figure 8. Cross-section of a typical Krem Puri passage developed within a single specific stratigraphic interval, with locations of rock samples. The column reports XRF analysis results (wt%) for Si, Al, Ca, and Mg (see Table 1) and for comparison average XRF results obtained for the Mahadek Sandstone sampled at surface near Mawsynram by Deka (2011).

Figure 9. Sample Fabric. A) KPs1X; B) KPs1E; C) KPs1A; D) KPs1B; E) KPs1C; F) KPs1D. See text for description.

Figure 10. SEM images of different mineral grains and weathering features in the sandstones of Krem Puri. A-B) Pitted quartz grain covered by apatite; C-D) K-feldspar with solutional pits; E-F) Albite crystal with solutional pits and associated kaolinite; G-H) Calcite crystal constituting the cement in KPs1c.

Figure 11. Analyses of heavy metals in water samples represented by bar lengths in milligram per litre. Electrical conductivity (EC), pH, and water discharge (Q) are indicated near the bars.

Figure 12. Trilinear diagram showing composition of water samples taken in Krem Puri.

Figure 13. Left: Total Dissolved Solids in water samples. Right: schematic 3-D representation of the vadose zone of the aquifer in which the cave develops, with hypothetical flow paths for water sampled in streams along cave galleries (circles) and drippings from fractures (drops). The water samples are listed on the left according to their increasing TDS content.

Figure 14. Left and centre) Cations and SiO_2 concentrations, and pH, are plotted against the total dissolved solid (TDS) content, where the increase in TDS is assumed to represent the geochemical evolution path of water in the studied aquifer. Right) Evolution of saturation indices of waters with respect to a selection of mineral phases.

Figure 15. A) Left: Plot of the relationships between the sum of Ca^{2+} and Mg^{2+} cations vs. HCO_3^-. The line for the ratio 1:1 (i.e. $[Ca^{2+}+Mg^{2+}] = 2[HCO_3^-]$) is represented. B) Right: Plot of the relationships Mg^{2+} vs. Ca^{2+}.

Figure 16. PHREEQC inverse modelling results (mmol/kg H_2O of transferred phases) for waters in Krem Puri aquifer, starting from water composition KPw8 and ending with water composition

KPw6 as in Fig. 13. The smaller black bars show the range of values obtained by three different models. Positive values are for dissolved phases and negative values are for precipitated ones. K-feldpsar (Kfs), albite (Ab), kaolinite (Kln), K-mica (Ms), illite (Ill), calcite (Cal), dolomite (Dol), gypsum (Gy), amorphous silica (SiO2(a)), CO_2.

Figure 17. Mineral stability relationships among Microcline K-feldspar, Kaolinite, Gibbsite, and Pyrophyllite in the presence of water at 25°C and 1 atm pressure. The dashed lines represent saturation with respect to quartz and amorphous silica.

Figure 18. Schematic evolution of the Krem Puri network. A) Phreatic speleogenetic phase when the carbonatic inception horizon was below the water table. B) Deactivation of the system due to the regional uplift, vadose/relict phase.