

HYDROGEOCHEMICAL EVOLUTION IN A TROPICAL CAVE SYSTEM; BATU CAVES, PENINSULAR MALAYSIA

NOR BAKHIAH BAHARIM^{1,*}, ROS FATIHAH MUHAMMAD² AND ISMAIL YUSOFF²

¹School of Marine and Environmental Sciences, Universiti Malaysia Terengganu, 21300 Kuala Nerus, Terengganu, Malaysia

²Department of Geology, Faculty of Science, University of Malaya, 50603 Kuala Lumpur, Malaysia

*Corresponding author: rosfmuhammad@um.edu.my

Abstract: A hydrogeochemical evolution of tropical caves' drip water was carried out at Batu Caves, Peninsular Malaysia. Drip water was analysed to underscore the cave hydrogeochemical evolution. The karst system of Batu Caves is characterized by low water storage capacity due to the high porosity of its weathered soil. A monitoring program was performed on a fortnightly time scale. The tropical environment showed a significant variation on the hydrogeochemical processes in the karst system compared to temperate environments. Drip water was assumed to be fed from a matrix of soil water flow in the absence of recharge. A constant hydraulic pressure was expected as a result of high evaporation in the tropical region and rainfall throughout the year. The mean drip rates varied from 0.02 to 1.02 ml/s and increased coincidentally with high water input, and vice versa. The drip water pattern suggested that the drip water differentiated in two flow regimes which are seasonal and seepage flow. Both regimes act differently and control the water infiltration process in the karst system. The water-rock interaction processes involved in the karst system are dissolution, dilution, ionic exchange and prior calcite precipitation (PCP) that lead to saturated $SI_{calcite}$ index. In general, drip water consists of Ca, Mg, and HCO_3 as dominant elements. The source rock deduction analyses strongly suggested that the hydrogeochemical properties of drip water originated from carbonate weathering. The drip and meteoric water were designated as Ca- HCO_3 and Na- CO_3 facies, respectively. Na ion in meteoric water replaced by Ca and SO_4 was exchanged with HCO_3 ; i.e., a Ca- HCO_3 as a result of the ionic exchange. The Ca and Ca:Mg determined that the dissolution rate declined as a result of the drip water reaching its saturation point against PCP. Most of the drip water reached oversaturated values ranging from 0.398 – 1.017. Each drip possessed a unique hydro-geochemistry characteristic which is significantly related to the host rock properties, flow path characteristic, fracture system behaviour, and volume of water input.

Keywords: hydrogeochemical, drip water, $SI_{calcite}$, ionic exchange, prior calcite precipitation

Introduction

In recent years, there has been a growing interest in understanding the hydro-geochemistry characteristics of karst systems (Fairchild *et al.*, 2000; Baker *et al.*, 2000; Tooth *et al.*, 2003; Baldini *et al.*, 2006; McDonald *et al.*, 2007; Karmann *et al.*, 2007). Predominantly, studies have been carried out in temperate regions such as the United Kingdom and Europe (Tooth *et al.*, 2003; Baldini *et al.*, 2006; Baker *et al.*, 2000; McDonald *et al.*, 2007; Fairchild *et al.*, 2006). These studies proved that the on-going processes form speleothem such as stalagmites preserve hydrological history records. These findings also provide new insight into the hydrogeochemical characteristics of

drip water as a paleoclimate proxy. Hydrology and geochemical characteristics of drip water are fundamentals of speleothem data proxies.

Extensive studies on drip water hydro-geochemistry revealed that significant factors act in karst systems, such as residence time of percolation water, prior calcite precipitation at individual drip site, volume of water input, flow routes taken at each drip sites, and the host rock composition (Baker *et al.*, 2000; Baldini *et al.*, 2006; Karmann *et al.*, 2007; Tooth & Fairchild, 2003). Detailed discussions mostly focused on the variability of Ca and Mg and their ratios that allowed the researcher to differentiate a rock water process in the karst system (Baker *et al.*, 2000; Baldini *et al.*, 2006; Karmann *et al.*,

2007; McDonald *et al.*, 2007; Tooth & Fairchild, 2003). The studies determined that the calcite saturation index ($SI_{calcite}$) increases during summer and decreases during winter. This is due to the low water storage in the karst system during low rainfall periods, thus triggering a proportion of ventilated air pocket reservoirs and derived degassing of CO_2 from the solution, causing calcite precipitation along the flow routes. Besides, a longer mean residence time of drip water favours the increment of $SI_{calcite}$ and enrichment as explained in Genty and Deflandre (1998) and Baker *et al.* (2000) who claimed that the increased conductivity is a result of the increment in water residence time in reservoirs. This phenomenon implies that the low water input during dry periods trigger a small amount of water accumulation in reservoirs and the hydraulic pressures become coincidentally low due to the slow water infiltration in the pores and microfissures of the system.

In addition, most of the previous studies highlighted that prior calcite precipitation (PCP) behaviour is a good indicator to determine the hydrogeochemical process in a karst system (Baker *et al.*, 2016; Baker *et al.*, 2000; Fairchild *et al.*, 2006). PCP is the occurrence of calcite precipitation in vadose zone water particularly to form speleothem due to the high/supersaturation of calcite concentration. This occurrence could indicate the residence time of water in the karst system based on the PCP amount. In addition, variations in the molar ratio of both these elements to Ca can be triggered by the process of prior calcite PCP, where $CaCO_3$ precipitates at upstream of the drip point due to CO_2 degassing into ventilated air pockets (Fairchild *et al.*, 2000).

Hydrogeochemistry acts in tropical region caves, especially in Peninsular Malaysia is still an open question since the very little study has been done. Previously, the hydrology of the karst system was studied by Crowther (1983). The study only focused on groundwater/seepage system surrounding the Setul Boundary Range, West Malaysia which comprised of Gua Tempurung, Perak, Gua Anak Takun,

Selangor and Perlis Mine Cave, Perlis. The findings concluded that the seepage discharge is mainly controlled by rainfall distribution with antecedent periods of 1-16 days. Therefore, the aim of this study is to prove the significant relationship between drip water and climatic parameters in this region. The expected outcome of this study is new results in terms of the physical and chemical properties of drip water, particularly in the case of Peninsular Malaysia.

The objectives of the study are to characterize drip water behaviour in tropical region caves and study their hydro-geochemistry. This preliminary study is a great base for interpreting the characteristics of drip water hydro-geochemistry in tropical region caves, especially in Peninsular Malaysia. The study may show that different processes may occur due to the relatively thin cover of karst hills in the tropical region and the high monsoonal rainfall throughout the year.

Study Area

The rainfall pattern in Peninsular Malaysia is influenced by the cycles of southeast monsoon, northeast monsoon, and inter-monsoon seasons. Batu Caves is located in the western part of Peninsular Malaysia at latitude $3^{\circ} 14' 16.6''$ and longitude $101^{\circ} 41' 04.8''$. It receives a high annual rainfall at an average value of 2937.5 mm.

The Batu Caves hill lies on the Kuala Lumpur limestone which comprises of a series of caves and cave temples (Figure 1). Batu Caves is illustrated as a tower karst that rises sharply with cliff-like surroundings and flat and karst edge plains in the tropics (Wycherely, 1971). A petrography study determined that the host rock of the Batu Caves hill is low-grade calcitic marble (Ng, 2004).

Villa Cave is situated at the foot of Batu Caves hill (Figure 2). It is 160.42 m long and the highest rooftop is about 43.89 m (Figure 3). The system is accessible and open to the public. The thickest limestone covers approximately

750 m. Generally, the cave exhibits inactive formation such as distinct and majestic notches and varieties of scallops. Only a few features of secondary deposits are observed in this cave such as stalactite and drapiers. All the features can be classified as young secondary deposits based on the size of the formations.

Dark Cave is located about 350 m above the base level and the thickness of its limestone

cover is approximately 400 m (Figure 3). The enclosed chamber of the Dark Cave at 45 m high and 2000 m provides animal habitats for bats, snakes, and guano feeders. This cave covers a wide range of secondary deposits such as stalagmites, stalactites, and flowstone. This cave is managed by the Malaysian Nature Society, which preserves its geomorphology. Description of each drip site is given in Table 1.

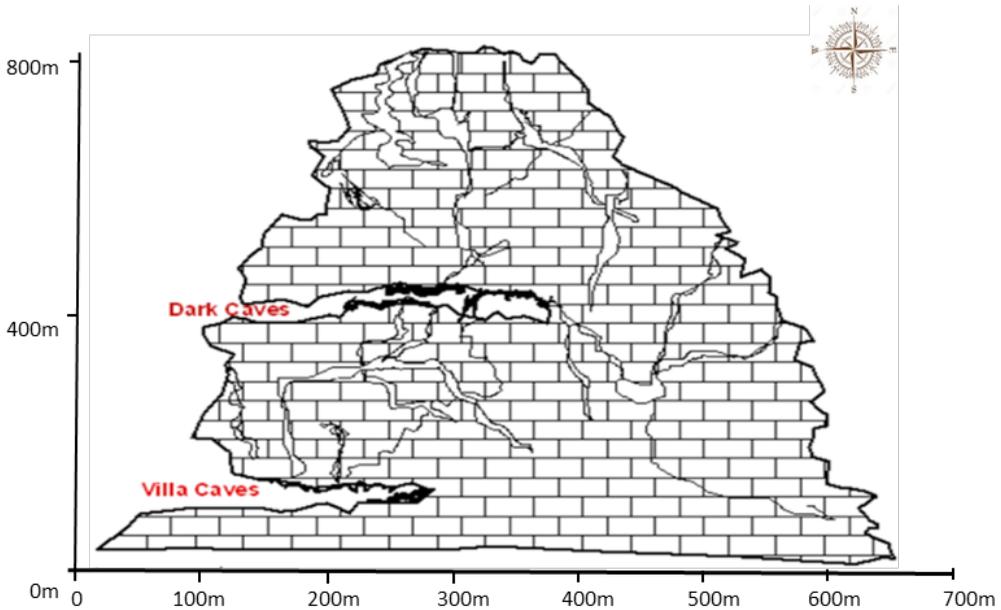


Figure 1: Hypothetical cross section of Batu Caves hill.

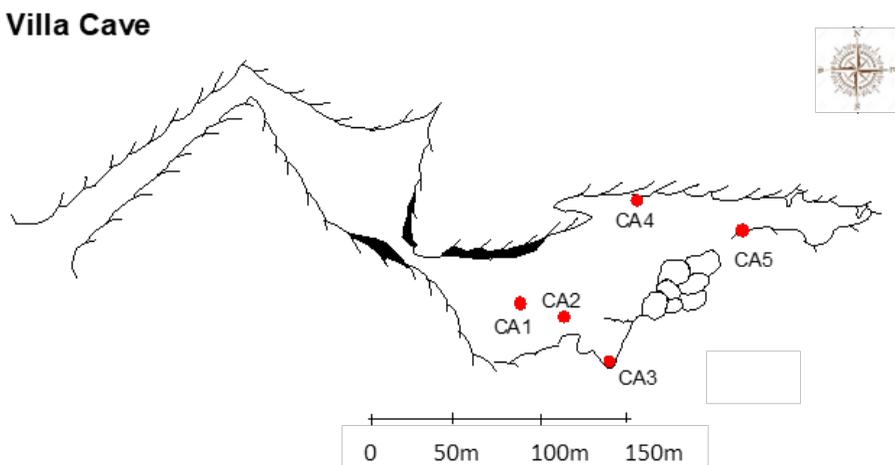


Figure 2: Map of Dark Cave and the sampling sites.

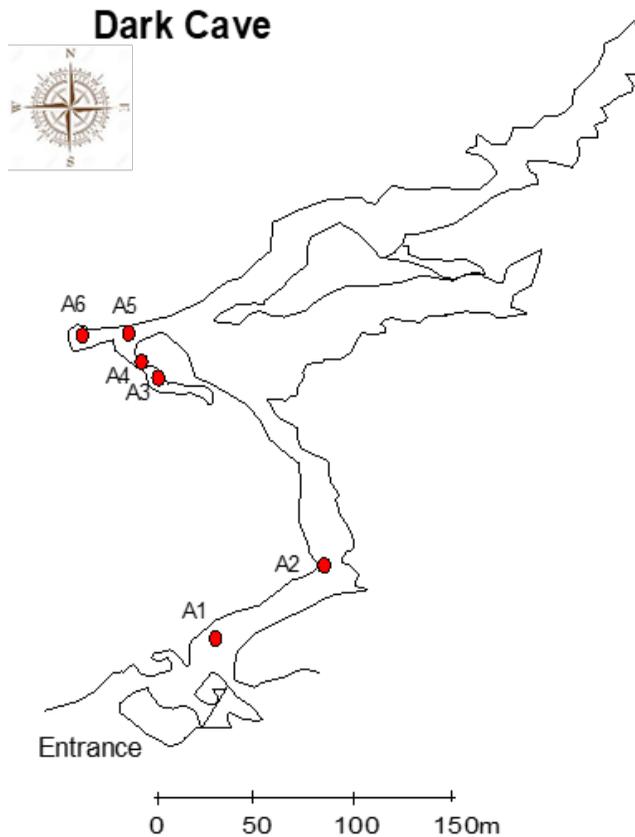


Figure 3: Map of Dark Cave and sampling sites.

Sample ID	Description
CA1	The drip water originated from small cracks in the cave ceiling.
CA2	The drip water emanated from drips onto small stalagmites.
CA3	The drips originated from small cracks and trickles through broken scallops.
CA4	The drip water emanated from the alluvium-covered ceiling.
CA5	Water flowed and trickled down from the wall covering.
A1	The drip water originated from draperies to form flowstone.
A2	The drip water emanated from a stalactite and drips onto a small stalagmite.
A3	The drips originated from drapery.
A4	Showerhead drip. The drip originated from a showerhead stalactite onto a pond.
A6	The drip water originated from a scallop ceiling.

Methodology

Sample collection was carried out fortnightly in Villa Cave and Dark Cave in October 2007-October 2008 and March 2008 – May

2008, respectively. Four samples of meteoric water were also collected at University Malaya (RKL), located about 10 km from the caves. In order to fulfil the objectives, the sites were chosen from different localities along the cave

passages. The sampling points were selected from stalactites, fracture openings, and ceilings. The discharge was measured using the stopwatch method to see how much time is taken for the drip water to accumulate in a 10 ml cylinder (McDonald et al., 2007).

All samples were collected using acid-washed low-density polyethylene (LDPE) bottles. The in-situ parameters measured consist of conductivity, total dissolved solid (TDS), dissolved oxygen (DO), pH, and temperature. Upon collection, the samples were divided into two aliquots. The samples were acidified using distilled HNO₃ (70%) to pH 2-4, then filtered through 0.45µm using a Nalgene™ pump filter. Acidified samples were analysed for cation species using inductively coupled plasma optical emission spectrometers (Perkin-Elmer). The second un-acidified aliquot was refrigerated prior to analyses for anion species (Cl⁻, SO₄²⁻, F⁻, and NO₃⁻) and alkalinity (HCO₃⁻ and CO₃²⁻) using Ion Chromatography and Auto-Titrator (Methrom). All laboratory analyses were carried out at the Department of Geology, University of Malaya.

All data were interpreted and presented using statistical analysis and graphical methods. The data analyses were conducted using Microsoft Office Excel and AQUACHEM software. The source rock deduction, *SI_{calcite}*, and Piper diagrams were evaluated using the PHREEQC program in the AQUACHEM software.

Results and Discussion

Physical and Chemical Properties

Table 2 summarizes the results of the physical and chemical parameters of the karst water. All drip sites maintain the flow throughout the year. Generally, the pH values ranged from 7.46 – 8.4, thus designating the place as a bicarbonate environment (Hounslow, 1995). Most water samples demonstrated TDS values below 500 mg/l which indicate silicate weathering, and is proportional to conductivity (Hounslow, 1995). However, water samples from the Villa Cave showed several episodes of greater TDS values exceeding 500 mg/l on 11/02/08 and 29/02/08. Overall, the results showed broad values of temperature ranging from 24.6 – 29.3 °C and the standard deviation was below 3. The dominant ions are displayed graphically in a Piper Diagram (Figure 4). The meteoric water had high NO₃ and Ca and the mean values were 2.17 mg/l and 1.381 mg/l, respectively. Besides, the results showed that the drip water had high Ca and NO₃ concentrations ranging from 24.28 to 78.75 mg/l and 6.85 to 70.84 mg/l, respectively. The drip water was designated as a Ca-HCO₃ facies as would be expected in a limestone environment. Similarly, meteoric water, RKL was assigned as a Na-SO₄ facies. The arrow in the Figure 4 suggests the alteration of Na-SO₄ facies (meteoric water) to Ca-HCO₃ facies (drip water) and shows the significant hydro-geochemistry evolution process acting in the karst system.

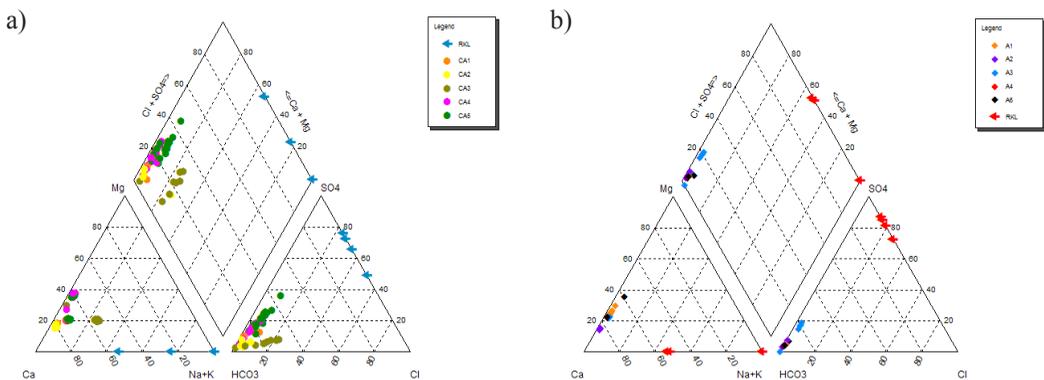


Figure 4: The Piper Diagram of drip water in Villa Cave, Batu Caves b: The Piper Diagram of drip water in Dark Cave, Batu Caves

Table 3 shows the results of the PHREEQC analyses which concluded that the drip water originates from carbonate weathering. The results also proved that Ca was derived from calcite and dolomite. However, the HCO₃: sum anions ratio showed that the drip water from Villa Cave (CA1, CA2, CA3, CA4, and CA5) and Dark Cave (A3) indicated brine (August 2008 and January 2008). Overall, the $SI_{calcite}$ ranged from being under-saturated to oversaturated with calcite. The results showed that 97% of the drip water in the Dark Cave has reached its oversaturated values ranging from 0.398 - 0.79. On the other hand, drip water in the Villa Cave showed that only 70% have reached oversaturated values ranging from -1.908 - 1.447.

definition grid of Smart and Friederich (1986) as modified by Baldini *et al.* (2006). Figure 5 shows that a positive log-linear correlation exists between the coefficient of variation (CV) and maximum discharge. The drop sites are divided into two classifications in terms of hydrological response to discharge which are seepage flow (CA5, A6, A4, and A3) and seasonal flow (CA1, CA2, CA3, CA4, A1, and A2). The mean value of drip rates ranged from 0.11 to 0.51 ml/s. Seepage flow was characterized as medium variable drips and the CV varied from 33.24 – 46.08. On the other hand, seepage flows demonstrated medium variability drip and constant flow. This suggests that the drip water flows through well-connected matrices and fractures. The well-connected matrices allow the water to consistently infiltrate through the reservoir with a constant hydraulic pressure.

Hydrology characteristics of the karst system

The mean discharge of drip sites in relation to the variation in discharge is plotted on the

Table 2: Mean value of physical and chemical parameters of drip water in Villa Cave (CA1-CA5) and Dark Cave (A1-A6).

Chemical Properties	CA1 n(5)	CA2 n(7)	CA3 n(11)	CA4 n(21)	CA5 n(29)	A1 n(6)	A2 n(6)	A3 n(6)	A4 n(6)	A6 n(6)
pH	7.12	7.29	7.36	7.8	7.84	8.07	7.82	8.00	8.09	7.95
Drip rates (mg/l)	0.51	0.17	0.05	0.07	0.11	0.51	0.17	0.05	0.07	0.11
Temperature (°C)	25.67	26.5	25.63	25.60	25.60	25.67	26.5	25.63	25.60	25.60
TDS (mg/l)	154.88	221.52	239.7	129.77	140.6	154.88	221.52	239.7	129.77	140.60
Conductivity (µS/cm)	321	456.33	491.83	267.8	292.5	321.00	456.33	491.83	267.8	292.5
Ca (mg/l)	67.89	48.93	33.54	51.43	50.28	44.13	71.39	58.07	40.60	43.58
Mg (mg/l)	9.24	6.31	8.25	16.88	9.45	9.56	7.58	10.33	4.72	8.48
Na (mg/l)	2.82	3.50	10.00	2.58	4.20	1.38	1.36	2.21	1.37	1.34
NO ₃ (mg/l)	35.66	35.01	42.52	64.45	70.84	12.42	6.85	70.21	6.63	10.56
HCO ₃ (mg/l)	219.96	187.22	139.78	164.94	132.1	159.49	232.94	134.75	126.8	153.47

Table 3: Source rock deduction analysis for drip water in Dark Cave (A1-A6) and Villa Cave (CA1-CA5), Batu Caves.

Parameter	Attention Value	Conclusion		A1 n(6)	A2 n(6)	A3 n(6)	A4 n(6)	A6 n(6)	CA1 n(6)	CA2 n(6)	CA3 n(6)	CA4 n(6)	CA5 n(6)	
Mg/ (Ca+Mg)	<0.5	Limestone-dolomite weathering	Min	0.247	0.142	0.221	0.148	0.318	0.1607	0.1771	0.319	0.3620	0.3675	
			Max	0.303	0.152	0.223	0.171	0.369	0.2162	0.2875	0.444	0.4912	0.4482	
			%	100%	100%	100%	100%	100%	100%	100%	100%	100%	38%	65%
	>0.5	Dolomite dissolution, calcite precipitation	Min	na	na	na	na	na	na	na	na	na	0.5021	0.5681
			Max										0.5629	0.6188
			%										62%	35%
TDS	>500	Carbonate weathering or brine	Min	81	125	119	70	90	843	80	na	513	63	
			Max	371	552	171	300	268	20%	0		516	480	
			%	100%	100%	83%	100%	100%				10%	91%	
	<500	Silicate weathering	Min	na	na	679	na	na	94	574	83	69	796	
			Max			17%			211		380	224	946	
			%						80%			90%	9%	
HCO3-/ Sum Anions	>0.8	Silicate or carbonate weathering	Min	0.86	0.91	na	0.854	0.832	0.835	0.825	0.98	na	0.344	
			Max	0.897	0.956		0.932	0.924	0.855	0.896	11%		0.779	
			%				100%	100%	40%	50%			100%	
	<0.8 sulfate low	Seawater or brine	Min	na	na	0.478	na	na	0.64	0.684	0.48	0.502	na	
			Max			0.757			0.78	0.781	0.739	0.791		
			%			100%			60%	50%	89%	100%		
SI Calcite	>0	Oversaturated with respect to calcite	Min	0.151	0.259	0.222	0.158	0.322	0.124	0.212	0.115	0.015	0.109	
			Max	0.666	0.867	0.797	0.797	0.657	0.904	1.01	0.477	0.906	0.578	
			%	100%	100%	100%	100%	63%	40%	50%	44%	90%	74%	
*na : no available	<0	Undersaturated with respect to calcite	Min	na	na	na	na	-0.398	1.447	-1.211	-1.908	-1.167	1.271	
			Max					33%	-0.192	-0.661	0.149	0.283	-0.042	
			%						60%	50%	56%	10%	26%	

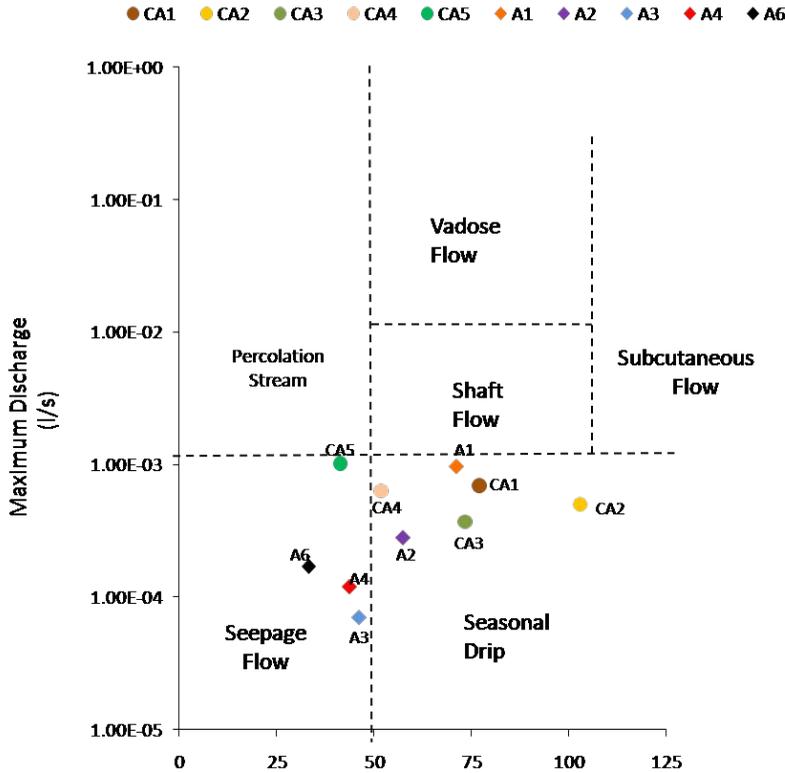


Figure 5: Maximum measured discharge for the drip rates plotted against the coefficient of variation (standard deviation/mean x 100). Adapted from Baldini *et al.*, (2006) after Smart and Friederich, (1986).

In addition, seasonal flow was characterized as high variability drips and responded intermittently with excess water at a CV range of 51.66 – 102.87. The seasonal flow indicated high variability drips with moderate discharge. This suggests that the seasonal flow intermittently responds to water input and is subject to underflow during large rainfall input flowing through poorly connected macro-pores and causing a high variable head of hydraulic pressure. The hydraulic pressure declined coincidentally with the decrease of water input. This implies a longer water residence time and allows sufficient time for water-rock interaction in the reservoir.

Figures 6 and 7 show the time series of drip rates, conductivity, and rainfall. Generally, the graph pattern shows that the drip rates and conductivity increased coincidentally with high rainfall events and vice versa. Overall, the pattern shows low water input in March and May of 2008 which triggered a small amount of water accumulation in the reservoir. Therefore, the hydraulic pressures became low as a result of slow water infiltration in the pores and microfissures, and the drip rates declined. In contrast, the drip rate was elevated in April due to rapid water infiltration during rainfall events which increased the hydraulic pressure in the karst system. Thus, the drip rates also increased.

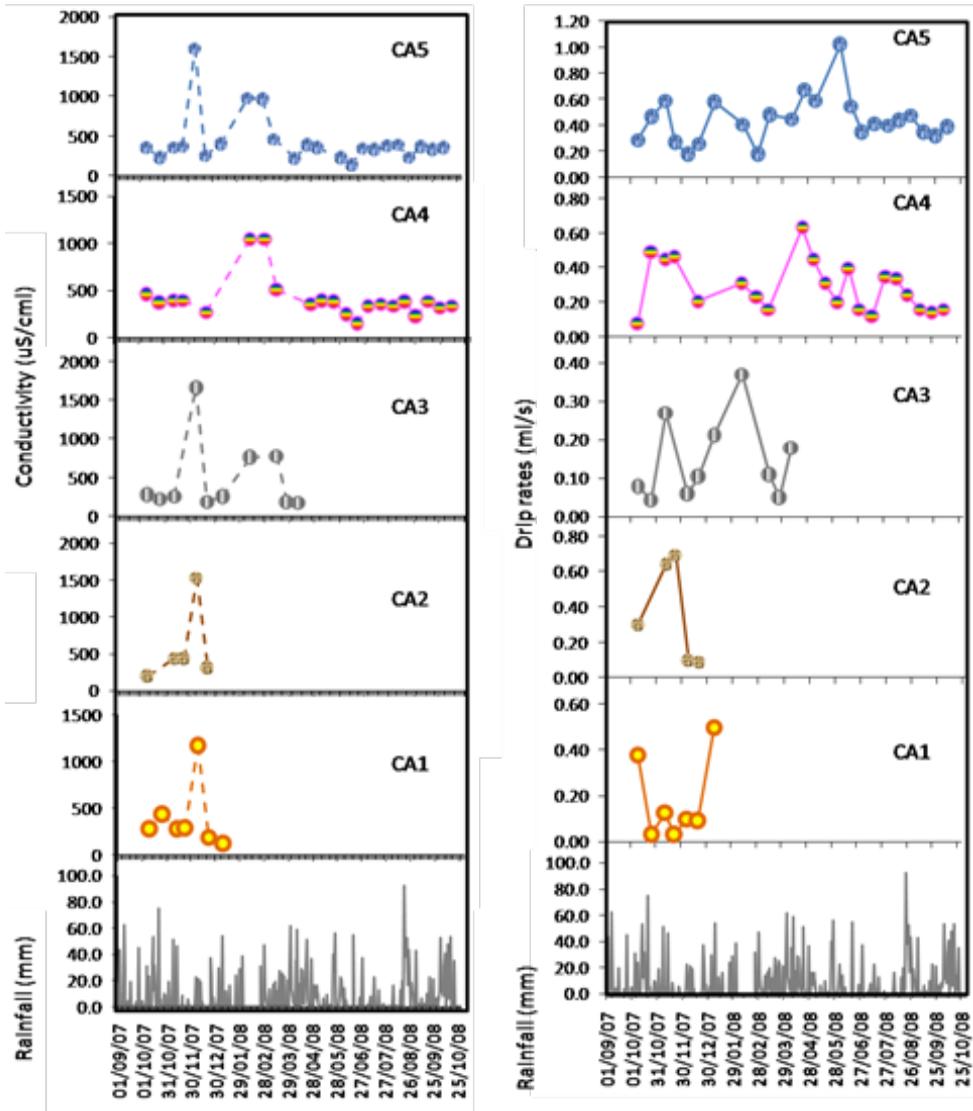


Figure 6: a) The conductivity and b) drip rates pattern of drip water in Villa Cave.

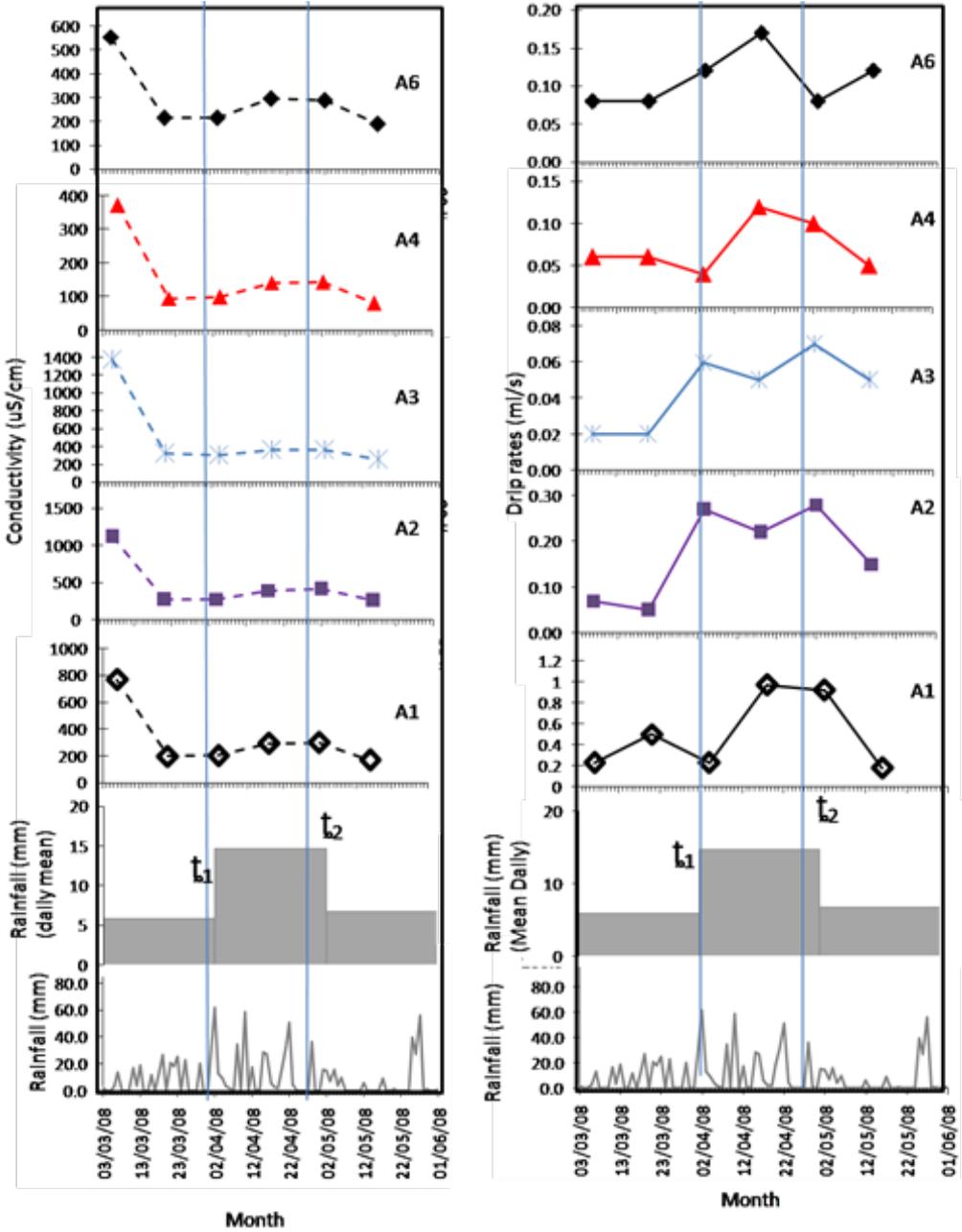


Figure 7: The a) conductivity and b) drip rates pattern of drip water in Dark Cave.

The time-lag indicates the filling/feeding time of water infiltrating through microfissures and pores in the karst (Fernandez-Cortes *et al.*, 2008). The findings show a sharp increase in drip rate and conductivity on 4/4/2008 (t_1) and 2/5/2008 (t_2) for drip sites A3 and A2. This could indicate that the time lag is short, varying from two to three days and showing that the drip rate corresponds to rainfall excess on 2/4/2008 and 30/4/2008. However, it should be noted that the time-lag determination was not accurately evaluated during water excess by considering the cave system flow path, therefore, the time-lag was proposed to be approximately less than one day. The time-lag study indicated a short time for water to fill in the karst system at approximately one day.

In comparison, previous studies have shown a range of drip site classifications (pages, seasonal, and subcutaneous flows) in the temperate region such as at Crag Cave, Ireland (Baldini *et al.*, 2006; Tooth & Fairchild, 2003) and Brown's Folly Mine (Fairchild *et al.*, 2006). This must be significant as different climate and topography characteristics change the hydrological properties of a cave system. In addition, a study monitoring the drip rates in the southern Spain revealed that the average filling time was 26 ± 10 days (Fernandez-Cortès, 2008). The moist environment in Peninsular Malaysia suggests that the hydraulic pressure is considered constant and stable throughout the year as a response to rapid water input. This suggests that each karst system is correspondingly complex responding to climatic conditions.

Geochemistry

The chemical properties demonstrated that the drip water is strongly influenced by the host rock composition. The source rock deduction analyses concluded that the karst water is derived from the limestone-dolomite weathering processes. The results showed an increase of TDS, Ca, and NO_3 in the Villa Cave. Since the Villa Cave is located in the upper parts of the Dark Cave and the limestone cover is around 750m, this suggests that the drip water in the

Villa Cave may experience long flow routes which increase surface reaction compared to drip water in the Dark Cave, thus coincidentally increasing the water-rock interaction process (Tooth & Fairchild, 2003). Furthermore, the high concentration of NO_3 could be derived from guano (Ford & Williams, 2007). This is strongly supported by the fact that Bullock (1972) identified approximately nine species of bats roosting in this caves and some parts of the cave floor were covered with thick guano. However, in the Villa Cave, the presence of bat habitats was only observed at the entrance. This suggests that the drip water in the Dark Cave reacted with guano and may directly infiltrate the Villa Cave and affected its chemical composition.

Despite the greater NO_3 content, the source rock deduction indicated that the HCO_3^- : sum anions ratio designated the water samples from the Villa Cave (CA1, CA2, CA3, CA4, and CA5) and the Dark Cave (A3) to contain brine which is impossible in a limestone environment. This could be due to the abundant NO_3 which originated from guano in this karst system which extensively altered the chemical composition of the karst water and may produce hydroxyapatite (Ford & Williams, 2007). The SI_{calcite} of the karst water from the Villa Cave was expected to be higher compared to the Dark Cave due to the increase of flow route travel distance which coincidentally increased the water-rock interaction process. However, the results showed that 97% of drip water in the Dark Cave was saturated but only 70% of drip water was saturated in the Villa Cave.

On the other hand, Figure 8 shows variations in Ca and Mg/Ca which showed a significantly negative correlation at each site and a parallel prior calcite precipitation of the PCP vector. The PCP behaviour is a good indicator to determine the hydrogeochemical process in the karst system. The results showed that Ca tend to decrease during PCP occurrences, thus increasing Mg. The trend showed that during high water input (high drip rates), the Mg/Ca ratios tend to decrease. This could be due to the dilution process which increased during

high water input as a result of new/young water present in the karst reservoir. Besides, Figure 9 shows that the dissolution rate declined as the solution tend to reach saturation with pH values above 6, proving that the PCP played a major role in this hydrogeological process. This is consistent with previous researches that claimed that the PCP tends to increase during summer coincidentally with low water input (Tooth et al., 2003; Baldini *et al.*, 2006). Figure 10 shows a negative correlation of Sclacite and drip rates. This implies that evapotranspiration increased during dry periods and triggered calcite precipitation.

There are significant differences in Ca concentrations among drip water sites and also between the drip water and other types of karst water. The factor affecting Ca variability is apparently the water-rock interaction processes

in the karst system. Differences in Ca are likely the result of varying degrees of PCP in the karst system, a mechanism that is very sensitive to hydrological changes in the karst system. Although a difference exists in the hydrological characteristics between drip sites, there is a remarkable consistency in the Mg:Ca ratios at both Villa Cave and Dark Cave (Figure 9). However, the Piper Diagram shows that CA3 is scattered from other drip sites (Figure 5). This suggests that the ionic exchange process plays a major role in the other drip sites and subsequently gives higher Ca and Mg. Besides, CA3 may experience a small potential of ionic exchange processes which increase the Na and SO_4 . This demonstrates that the variance of the flow path in the karst system and characteristics influenced the chemical properties of drip water.

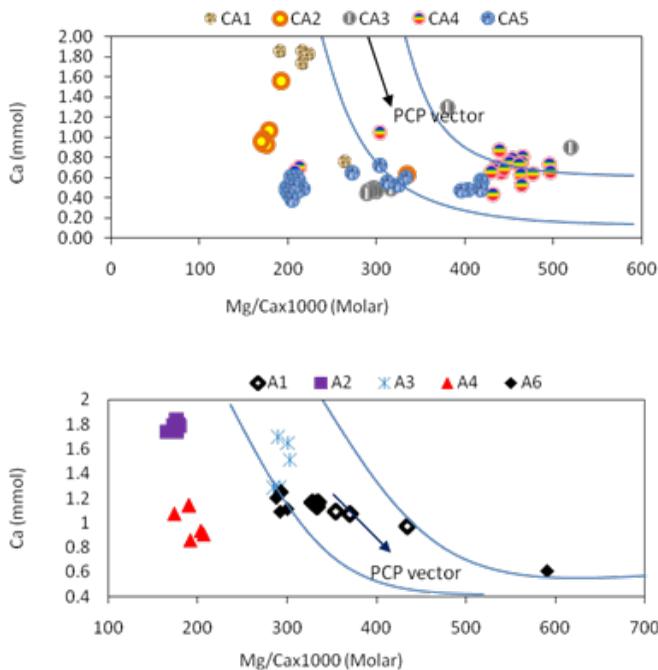


Figure 8: Ca against Mg:Ca to isolate the effects of calcite precipitation.

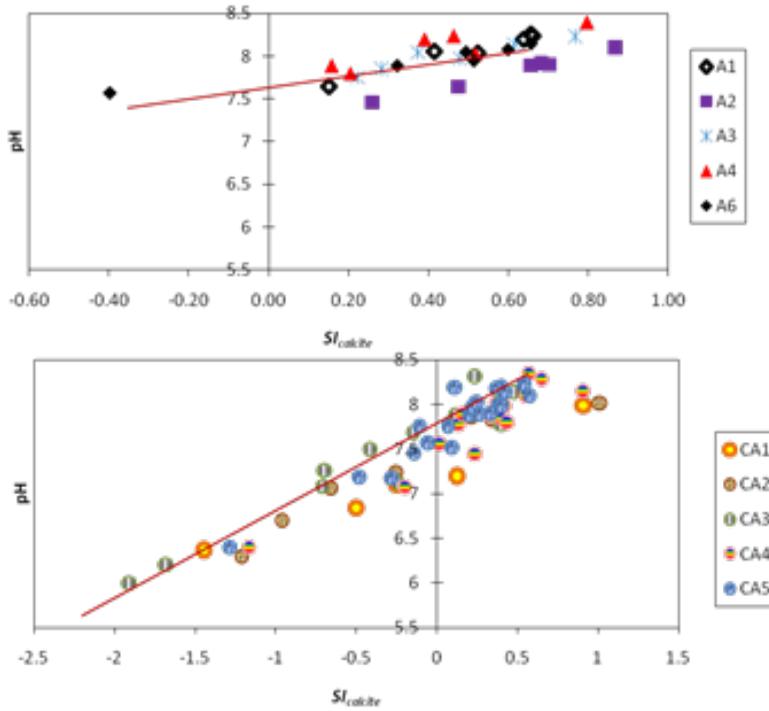


Figure 9: The relationship of pH value and $SI_{calcite}$.

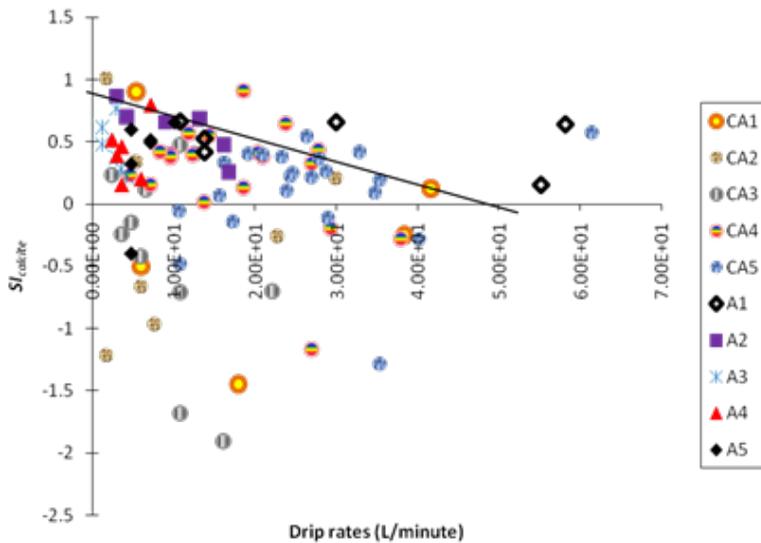


Figure 10: The correlation of $SI_{calcite}$ ratios and drip rates

Batu Caves' Hydrogeochemical Evolution

Figure 11 shows a conceptual model of hydrogeochemistry evolution in karst system. The figure shows that in the intermediate zone, the dissolution and dilution processes dominantly control the karst system. The processes increased during high water input as a result of young water infiltrating through the matrix soil and fractures. The meteoric water infiltrating through the intermediate zone showed a significant evolution of Na-SO₄ facies to Ca-HCO₃ facies. The evolution implies a significant ionic exchange process in the karst system. The Na ion was replaced by Ca, and SO₄ was exchanged with HCO₃ to produce Ca-HCO₃.

Water input and drip rate monitoring indicated that the drop sites can be classified into two groups based on their hydrological responses which are medium variable drip (seepage flow) and high variable drip (seasonal flow). The seepage flow and seasonal flow conveniently identify the end-members in a spectrum of possible flow regime types. The drip rate tends to increase with high rainfall (high water input) events, and vice versa. The

volume of water input controls the hydraulic pressure and delivery potential of water infiltrating through the lower zone. Then, PCP plays a dominant process in the lower zone.

The $SI_{calcite}$ value showed that most of the drip water was saturated with calcite. Also, the high pH values showed a significant decline of dissolution rate in the zone. The high $SI_{calcite}$ value in the Dark Cave designated the potential of speleothem deposition. This is proven by the fact that the Dark Cave is extensively covered with speleothems such as stalagmites, stalactites, and ponds, thus proving the growth of secondary deposits.

In addition, the flow routes and the travel distance of drip water must be taken into account for the control of the hydrogeochemistry evolution in the karst system. The variations in limestone cover of the Dark Cave and Villa Cave caused the difference in chemical properties of drip water. Moreover, the individual characteristics of flow routes are also a strong factor controlling the mechanism of hydro-geochemistry processes in the karst system.

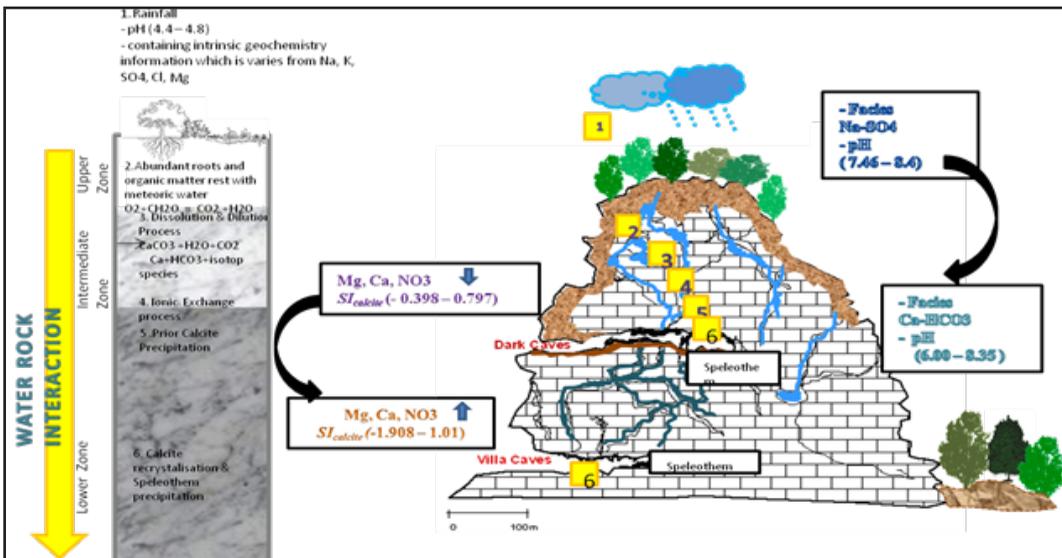


Figure 11: The conceptual model of hydrogeochemistry evolution in karst system.

Conclusion

Generally, the physical and chemical properties revealed the behaviour of drip water. A preliminary study showed a significant relationship between the volume of water input and the hydrological characteristic of the karst system. Throughout the year, all drip sites maintained flow, which can be classified into seasonal and seepage flows. Furthermore, the study also revealed that the filling time of water infiltrating through pores and microfissures is approximately less than three days. This study highlighted that the hydraulic pressures play a major role in the karst hydrological system where the hydraulic pressures increased coincidentally with high rainfall events.

Even though the major elements of drip water are Ca and Mg, each drip site showed unique hydro-geochemistry characteristics related to the host rock properties, flow path characteristics, fracture system behaviour and volume of water input. The conceptual model simplified the hydrogeochemical processes in the karst system. Overall, the processes of dilution, dissolution, ionic exchange, and prior calcite precipitation within the fracture would determine the evolution and water chemistry of drip water.

In conclusion, this study proved the significant relationship between drip water and climatic parameters. However, the understanding of implications from the climatic record is limited by the temporal limitation of monthly sampling. Overall, this study pointed towards the critical importance of high-frequency sampling of drip water and continuous monitoring of drip rates. This strategy will provide more detailed information regarding the processes in a karst system. In addition, a combination of element abundance and isotopic study would be a powerful approach.

Acknowledgements

The authors are grateful to the staff of the Department of Geology, University of Malaya,

for the water sample analysis. Special thanks to late Prof. Hutchison for their ideas and valuable suggestions. This study was funded by the Ministry of Science, Technology and Innovation, Malaysia.

References

- Baker, A., Flemons, I., Andersen, M. S., Coleborn, K., & Treble, P. C. (2016). What determines the calcium concentration of speleothem-forming drip waters? *Global and Planetary Change*. 143: 152–161.
- Baker, A., Genty, D., & Fairchild, I. J. (2000). Hydrological characterisation of stalagmite dripwaters at Grotte de Villars, Dordogne, by the analysis of inorganic species and luminescent organic matter. *Hydrology and Earth System Sciences*. 4(3): 439–449
- Baldini, J. U. L., McDermott, F., & Fairchild, I. J. (2006). Spatial variability in cave drip water hydrochemistry: Implications for stalagmite paleoclimate records. *Chemical Geology*. 235(3): 390–404.
- Bullock, J. A. (1972). *Malayan Nature Journal. Cave Biology in Malaysia*. 25: 135–141.
- Crowther, J. (1983). Hydrology of autogenic percolation systems in some tropical karst outcrops, West Malaysia. *Journal of Hydrology*. 60(4): 227–242.
- Fairchild, I. J., Tuckwell, G. W., Baker, A., & Tooth, A. F. (2006). Modelling of dripwater hydrology and hydrogeochemistry in a weakly karstified aquifer (Bath, UK): Implications for climate change studies. *Journal of Hydrology*. 321(1): 213–231.
- Fernandez-Cortès, A., Calaforra, J. M., & Sánchez-Martos, F. (2008). Hydrogeochemical processes as environmental indicators in drip water: Study of the Cueva del Agua (Southern Spain). *International Journal of Speleology*. 37(1): 41–52.
- Ford, D., & Williams, P. (2007). *Karst*

- Hydrogeology and Geomorphology*. John Wiley & Sons.
- Hounslow. (1995). *Water Quality Data: Analysis and Interpretation*. Lewis Publishers.
- Karmann, I., Cruz, F. W., Viana, O., & Burns, S. J. (2007). Climate influence on geochemistry parameters of waters from Santana-Pérolas cave system, Brazil. *Chemical Geology*. 244(1): 232–247.
- McDonald, J., Drysdale, R., Hill, D., Chisari, R., & Wong, H. (2007). The hydrochemical response of cave drip waters to sub-annual and inter-annual climate variability, Wombeyan Caves, SE Australia. *Chemical Geology*. 244(3): 605–623.
- Ng, P. P. (2004). *Kajian petrografi batu kapur kawasan Batu Caves, Selangor Darul Ehsan*. Universiti Malaya.
- Tooth, A. F., & Fairchild, I. J. (2003). Soil and karst aquifer hydrological controls on the geochemical evolution of speleothem-forming drip waters, Crag Cave, southwest Ireland. *Journal of Hydrology*. 273(4): 51–68.
- Wycherely, P. (1971). *History of Batu Caves: In: Soepadmo E and Ho Thian Hua (ed), A Guide to Batu Caves*. Nature Society.