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Spatio-temporal variation of karst spring parameters for characterizing of the aquifer system of Watuputih Area, Indonesia

Taat SETIAWAN1), 2) ✉, **Boy Y.C.S.S. SYAH ALAM**2),
Eko HARYONO3), **HENDARMAWAN**2)

1) Geological Agency of Indonesia, Ministry of Energy and Mineral Resources, Indonesia
2) Padjadjaran University, Faculty of Geological Engineering, Jatinangor 45363, Sumedang, Indonesia
3) Gadjah Mada University, Faculty of Geography, Bulaksumur 55281, Yogyakarta, Indonesia

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Abstract

There are several springs with the large discharge around the Watuputih Karst Hills area that playing a crucial role in supplying water for both domestic and irrigation needs. The springs are located in the fault and fold zones of the Rembang anticlinorium system. This study was designed to determine the characteristics of karst aquifers from one year of monthly spatio-temporal data on discharge parameters and physico-chemical properties (temperature, pH, *EC*, Ca^{2+}, Mg^{2+}, HCO_3^-) of the four major springs, namely Brubulan Tahunan, Sumbersemen, Brubulan Pesucen, and Sendang Sayuran. It used statistical calculations to characterize spring discharge and hydrochemical variations, as well as bivariate correlation analysis and flow-duration curve (*FDC*). The variability index (I_v), variability (*V*), and spring coefficient of variation parameters (*SCVP*) classified Brubulan Tahunan and Sumbersemen as springs producing stable, fairly constant discharge with low variations but characterized Brubulan Pesucen as having unstable, varying discharge with moderate variations. The results showed gently sloping hydrograph, low variations in discharge and hydrochemical properties, a relatively prolonged response of discharge and CO_2-H_2O-$CaCO_3$ interaction to rainfall, and slope changes in the FDC. In other terms, although the springs are controlled by faults and folds, they have diffuse groundwater storage system in the form of densely fractured and porous media. These findings also indicate a less developed interconnected conduit, although Brubulan Pesucen is relatively more developed than Sumbersemen and Brubulan Tahunan. The geological structure and hydraulic gradient formed between the groundwater recharge and discharge areas are proven to control the amount of spring discharge actively.

Key words: *flow system, hydrochemical properties, Indonesia, karst aquifer, spring variability*

INTRODUCTION

Karst regions have specific hydrogeological characteristics because their constituent rocks, including limestone and dolomite, are prone to chemical dissolution [FORD, WILLIAMS 2007]. Karst aquifers have complex characters with groundwater flowing through three media or systems, namely pores, fractures, and conduit [GOLDSCHEIDER, DREW (ed.) 2007]. While groundwater in conduit systems has a relatively short residence time, the one in densely fractured or diffuse systems has a long one [BAENA *et al.* 2009]. Identifying groundwater flow systems in karst aquifers requires more detailed observations than in non-karst systems that have more stable responses [OZYURT, BAYARI 2008]. Spring hydrographs and chemographs are forms of temporal data analysis to identify the characteristics of karst groundwater flow [RAEISI, KARAMI 1997], which reflect the responses of aquifers to groundwater recharge processes [FORD, WILLIAMS 2007]. Variation in discharge amount and physico-chemical water properties can also be used to determine groundwater recharge areas, groundwater flow rates, and geographical conditions of an area [SHUSTER, WHITE 1971].

There have been previous studies on the characteristics of karst groundwater flow based on temporal variations in discharge parameters and the physico-chemical properties of springs, for instance, FOURNIER *et al.* [2007], BAENA *et al.* [2009], and PETRIČ, and KOGOVŠEK [2010]. FOURNIER *et al.* [2007] have successfully identified the mechanism of groundwater flow and phosphate contamination through the conduit system and nitrate contamination through the diffusion mechanism. Based on water temperature, BAENA *et al.* [2009] have identified a high variation in groundwater flow properties in conduit systems and low variation in diffusion systems. PETRIČ and KOGOVŠEK [2010] confirm that the fluctuation of air temperature affects groundwater through conduit systems. Also, the characteristics of karst groundwater flow in Indonesia, especially in Gunungsewu, have been analyzed from spatio-temporal variation in spring hydrograph and hydrochemistry, e.g., ADJI [2012] and ADJI *et al.* [2016]. These studies can generally explain the groundwater flow profiles during the processes of recharge, drainage, storage, and emergence in springs. Groundwater flow is related to the hydrochemical characters and karstification degree of an area. Diffusion flow is associated with undeveloped karstification and typified by relatively high dissolved ions. It is dominant in the absence of flood and during flow recession.

This paper explains the results of a one-year spatio-temporal study of the physico-chemical water parameters in four major springs in the Watuputih Hills and their surroundings, which is an attempt to recognizing the karst aquifer characteristics. This area is located in Rembang Regency, Central Java Province, Indonesia and is a limestone mining site in the Paciran Formation. Since 2014, the area has been widely discussed following an increasing public concern about the sustainability of the springs due to limestone mining. For these reasons, a thorough understanding of the aquifer characteristics of the major springs can necessarily provide a basis for spring management [KATSANOU *et al.* 2015]. This paper focuses on four springs, namely Brubulan Tahunan, Sumbersemen, Brubulan Pesucen, and Sendang Sayuran whose emergences are controlled by geological structures, namely faults and folds [GAI 2017]. Except for Sendang Sayuran, these springs have large discharges that essentially supply the domestic and irrigation needs of the public.

In the characterization of karst aquifers, this study is included in the preliminary stage. Compared to previous research, it offers a novelty in describing the features of springs emerging from the complexity of the regional geological condition, which is stratigraphy with alternating karst and non-karst rocks, and structures that cut through and fold these rocks. All of which allow the main springs in the study area to have a storage system from not only karst but also non-karst aquifers and enable the transfer mechanism between the two types of aquifers through the geological structures above.

MATERIALS AND METHODS

GEOLOGICAL AND HYDROGEOLOGICAL SETTING

Physiographically, the study area lies in the Rembang anticlinorium zone that is characterized by hills with steep to gentle slopes at an elevation of averagely less than 500 m a.s.l. [BEMMELEN 1949]. Geologically, this area consists of various rock formations, including carbonate rocks, siliciclastic sediments, and volcanic rocks [LUTHFI *et al.* 2017; NOVITA *et al.* 2017]. The karst hills have developed into isolated cone patterns in the limestones of the Bulu and the Tawun Formations, while the limestone in the Paciran Formation has not shown any morphological development (Fig. 1).

The study area is composed of several formations of limestone and sandstone from the Early Miocene to Pleis-

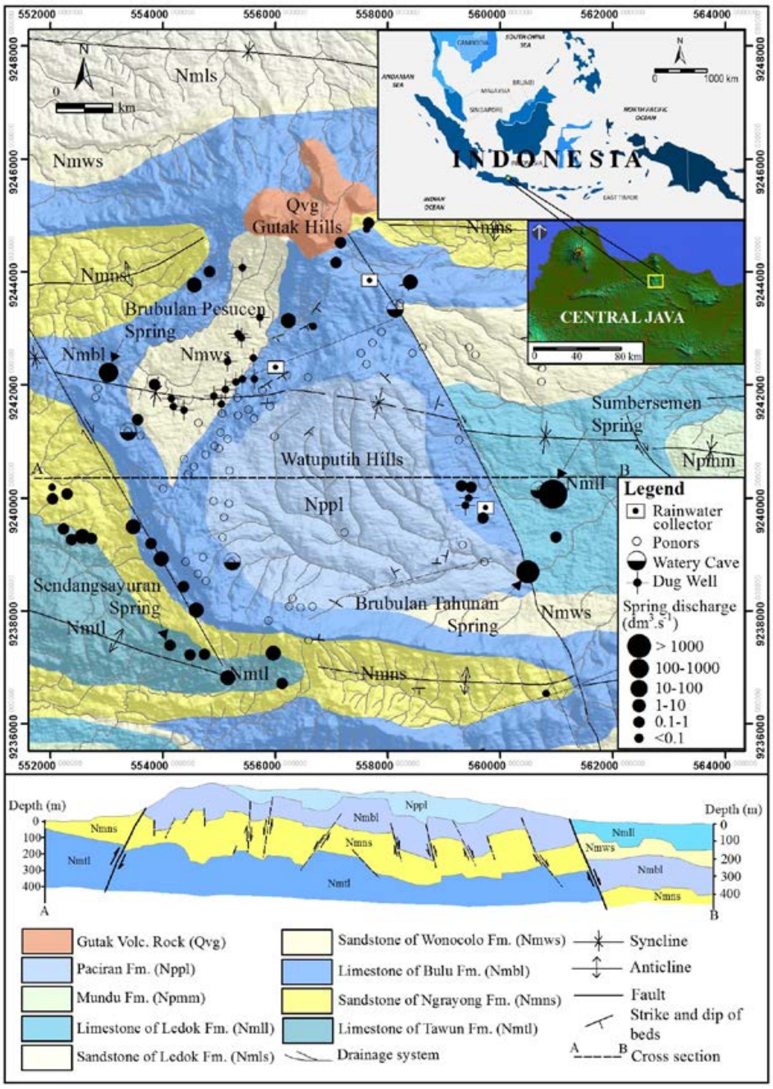

Fig. 1. The map of geology and hydrogeological objects of the study area; source: own elaboration

tocene [LUTHFI *et al.* 2017; NOVITA *et al.* 2017]. Formed in the upper of Early Miocene, the limestone of the Tawun Formation (Nmtl) is the oldest rock in the local stratigraphy. It is composed of sandy marl alternating with bioclastic limestone (packstone-wackestone). The quartz sandstone of the Ngrayong Formation (Nmns) from the Middle Miocene comprises quartz sandstone with limestones and sandy limestones intercalations. The limestone of the Bulu Formation (Nmbl) was formed in the upper of the Middle Miocene and is composed of bedded clastic limestone that is solid, locally jointed, and porous. The sandstone of the Wonocolo Formation (Nmws) is the upper of Middle Miocene and consists of calcareous sandstone with sandy marl intercalations. The sandstone of the Ledok Formation (Nmls) from the Late Miocene is composed of glauconitic sandstones cemented by carbonates. The limestone of the Ledok Formation (Nmll) was formed in Late Miocene and is composed of clastic limestones (wackestone-packstone). The marl of the Mundul Formation (Npmm) is of the Pliocene epoch and is distributed around the synclinal axis.

The limestone of the Paciran Formation (Nppl), formed from the Pliocene to Pleistocene, is composed of clastic limestones (packstone-wackestone) and can be found in the Watuputih Hills. The Gutak Volcanic Rocks in the North of the study area (Qvg) have the age of Pleistocene and consist of dacitic andesitic lava, andesitic breccia, and andesitic intrusions. The developing geological structures are faults, folds (syncline, anticline), and lineaments – both anticlines in the South and North plunge into two directions. The syncline in the middle also plunges into two directions, while the one in the North plunges from the East. The faults in the East and West, stretching with the NW-SE direction, are strike-slip faults [LUTHFI *et al.* 2017; NOVITA *et al.* 2017]. Geophysical investigation using resistivity and gravitational anomaly indicates the presence of the downthrown part in the eastern strike-slip fault and a slightly upward displaced element of the western strike-slip fault [GAI 2017].

The hydrogeological systems involve the emergence of 39 springs and four watery caves in the limestone of the Paciran Formation (Fig. 1). These springs emerge from fault zones, fold systems, and contacts between karst and non-karst rocks [GAI 2017]. The study focuses on four major springs that have a significant role in the surrounding localities, namely Sumbersemen, Brubulan Tahunan, Brubulan Pesucen, and Sendang Sayuran. Based on the karst spring classification proposed by FORD and WILLIAMS [2007], they fall into the category of artesian-fault guided springs that appear through a fault plane or an eroded impermeable layer on fold limbs (Tab. 1).

FIELD SAMPLING AND MEASUREMENT OF SPRINGS DISCHARGE

For hydrochemical analysis, the water from the four springs was sampled from January 2018 to January 2019 (except June and November). Some parameters were measured in the field, including temperature (T), degree of acidity (pH), electric conductivity (EC), and concentration of HCO_3^-. The T, EC, and pH were measured with the Lamotte Test Kit Portable, while the concentration of HCO_3^- was assessed by titration with the Alkalinity Test Kit. The water sample was filtered using a 0.45 µm syringe filter and then put into a 200 cm^3 polyethylene bottle each for anion and cation. Meanwhile, for cation analysis, each water sample was acidified with 0.1 N HNO_3 to prevent precipitation. Before the laboratory analysis, all water samples were preserved at 4°C. Also, the rainfall intensity was measured at three locations around the study area.

The spring discharge was calculated by multiplying flow velocity by wet cross-sectional area that had been divided into several horizontal segments with a particular distance. In this research, the water flow velocity was measured using a current meter. The springs discharge was measured at location that met the following criteria: straight channel, stable flow, no turbulence, no backwater effects, and no aquatic plants.

LABORATORY ANALYSIS

Major ions, including Ca^{2+}, Na^+, Mg^{2+}, K^+, HCO_3^-, SO_4^{2-}, Cl^-, and NO_3^-, were analyzed in the hydrochemical laboratory of the Center for Groundwater and Environmental Geology, the Geological Agency of Indonesia (Ind. Pusat Air Tanah dan Geologi Tata Lingkungan, Badan Geologi). Ca^{2+}, Na^+, Mg^{2+}, and K^+ were analyzed using the Dionex ICS-1500 Ion Chromatography System, while SO_4^{2-} and NO_3^- were assessed with a Varian Cary 100 UV-Visible Spectrophotometer. Cl^- and HCO_3^- were titrated with argentometry and alkalinity, respectively. The laboratory analysis results are applicable only if their charge balance error (CBE) is less than 5% [YUAN *et al.* 2017].

Table 1. The geological conditions of the springs in the study area

Spring	Elevation (m a.s.l.)	Geological structure	Lithology	Notes
Brubulan Tahunan	155	normal strike-slip fault	fault contact between the limestone of Bulu Formation (Nmbl) and the limestone of Ledok Formation (Nmll)	artesian spring
Sumbersemen	155	on a synclinal limb, a fault underneath the spring	the limestone of Ledok Formation (Nmll).	the largest discharge and artesian spring
Brubulan Pesucen	237	close to a synclinal axis	the limestone of Bulu Formation (Nmbl).	horizontal cavities on the spring outlet
Sendang Sayuran	288	close to the axis of a plunging syncline	the limestone of Tawun Formation (Nmtl)	the smallest discharge

Source: own study.

SATURATION INDICES AND PARTIAL PRESSURE OF CO$_2$

Mineral saturation indices (*SI*) and partial pressure of CO$_2$ (P_{CO2}) significantly define the characteristics of karst aquifer systems [White 2015]. The P_{CO2} is the basis for identifying the level of water-CO$_2$ interaction using the equation below [Ford, Williams 2007]:

$$Pco_2 = \frac{(HCO_3^-)\,(H^+)}{K_1\,K_{CO_2}} \tag{1}$$

Where (HCO_3^-) is bicarbonate ions, (H^+) is hydrogen ion activities, K_1 is the equilibrium reaction constant of solution at 25°C, and K_{CO2} is the equilibrium constant of CO$_2$ in the water at a temperature of 25°C.

Mineral saturation indices (*SI*) represents the chemical equilibrium between water and constituent minerals of aquifers [Ford, Williams 2007; Yuan *et al.* 2017], which can be calculated using the equation below:

$$SI = \log IAP : K \tag{2}$$

Where K is the mineral equilibrium constant, and *IAP* is the ion activity product of the mineral. $SI > 0$ represents mineral saturation, $SI < 0$ marks mineral undersaturation, and $SI = 0$ suggests equilibrium in water-rock interaction. The *SI* was calculated from the saturation of calcite (CaCO$_3$), expressed as SI_C. The SI_C values generally have a margin of error of ±0.1–0.2 [Ford, Williams 2007]. The mineral saturation and partial pressure of CO$_2$ (P_{CO2}) were calculated using the PHREEQC software [Parkhurst, Appelo 1999].

STATISTICAL ANALYSIS

The statistical analysis employed in this study included the characterization of spring discharge, bivariate analysis of hydrochemical parameters, and flow duration curve analysis. Each of these analyses is explained as follows.

Spring discharge changes at specific amplitude and frequency, which are attributable to the geometry and physical properties of the aquifer. Therefore, in spring management, characterization is fundamental [Malik 2015]. Variability index (I_v) classifies springs based on variations in the amount of discharge – which, according to Malik [2015], is the ratio of maximum discharge (Q_{max}) to minimum discharge (Q_{min}).

$$I_v = \frac{Q_{max}}{Q_{min}} \tag{3}$$

According to the Slovak Hydrometeorological Institute [Malik 2015], the stability of spring discharge and its corresponding range of variability index (*Iv*) are classified as stable (1.0–2.0), unstable (2.1–10.0), very unstable (10.1–30.0), and totally unstable (>30.0).

Meinzer [1923] acc. to Malik [2015] proposes a more detailed characterization from the variability value of the spring, as formulated below.

$$V = \frac{Q_{max} - Q_{min}}{Ø}\ 100\ (\%) \tag{4}$$

Where: V denotes variability index of spring in %, Q_{max} and Q_{min} are maximum and minimum discharge, and Ø is the arithmetic average of spring discharge. $V < 25\%$ represents a constant spring, while $V > 100\%$ defines spring as variable or fluctuating.

Flora [2004] and Springer *et al.* [2004] classify springs based on the spring coefficient of variation parameter (*SCVP*), which can be computed with the mathematical equation below.

$$SCVP = \frac{\sigma}{Ø} \tag{5}$$

Where: σ is the standard deviation of spring discharge and Ø is the arithmetic average of spring discharge.

According to Flora [2004] and Springer *et al.* [2004], the classification of spring variability based on *SCVP* is classified as low (0–49), moderate (50–99), high (100–199), and very high (>200).

The hydrochemical parameters were processed using the bivariate analysis. This statistical function produces a coefficient of correlation (R) by measuring the direction and strength of the linear line formed of two variables [Kovacs *et al.* 2012], as formulated below:

$$R_{(x,y)} = \frac{\sigma_{(x,y)}}{\sigma_x \sigma_y} \tag{6}$$

Where: σ_x, σ_y are the standard deviations of x and y variables and $\sigma_{(x,y)}$ is covariance.

Flow-duration curve (*FDC*) is a graph that displays the relationship between flow discharge and its exceedance probability [Katsanou *et al.* 2015]. *FDC* can distinguish the hydraulic characteristics, aquifer geometry, and groundwater recharge systems of one spring from another [Malik 2015]. *FDC* analysis was carried out by referring to the methodological approach described by Katsanou *et al.* [2015], which characterize the springs in a complex karst system in the Louros Basin in northwestern Greece. As for the spring discharge probability, this data was computed in statistical software, SPSS v.20.

RESULTS

DISCHARGE VARIATION AND SPRING CLASSIFICATION

The results of monthly discharge measurements at Brubulan Tahunan, Sumbersemen, and Brubulan Pesucen Springs from January 2018 through January 2019 *(*except June and November) are presented in Figure 2. The discharge of Sendang Sayuran Spring was not measurable because its water was directly stored and distributed publicly. However, the measurement in April 2017 showed that it had an instantaneous discharge of 5 dm^3·s^{-1} [GAI 2017]. The discharge measurements at the remaining three springs showed different variations. Brubulan Tahunan Spring released water at 127.42 m^3·s^{-1} in October and up to 199.82 dm^3·s^{-1} in March (average = 157.73 dm^3·s^{-1}). Sumbersemen Spring produced water at a varying discharge from 1322.22 dm^3·s^{-1} in December to 1916.67 dm^3·s^{-1} in May (average = 1584.63 dm^3·s^{-1}), while the discharge at

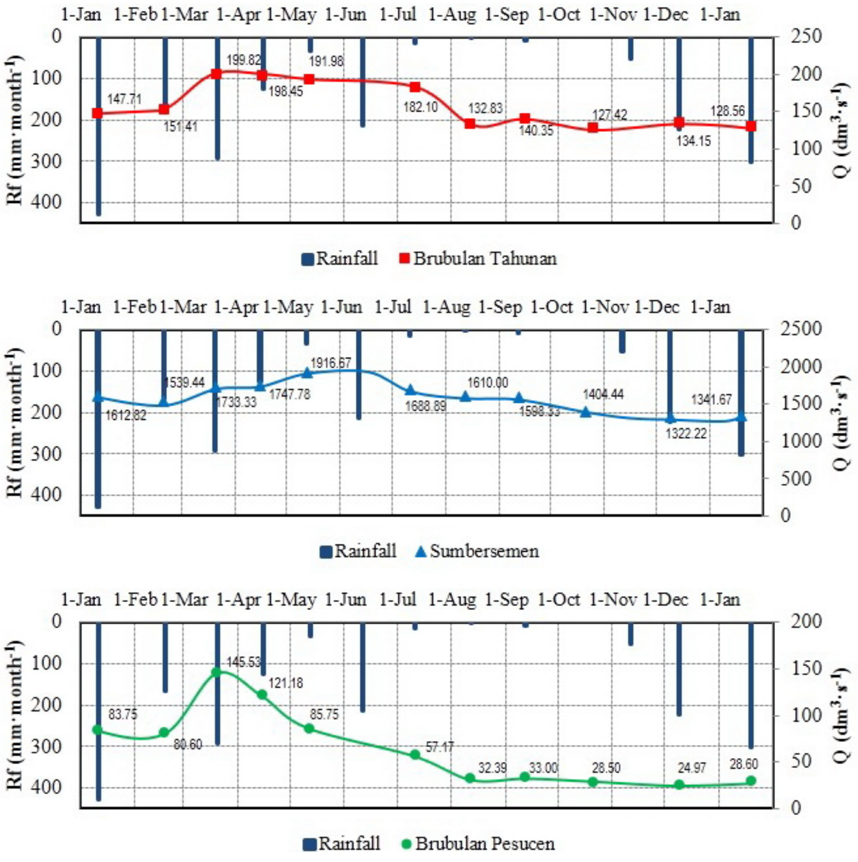

Fig. 2. Graph of monthly rainfall (*Rf*) and spring discharge (*Q*) variations at three major springs in the study area; source: own study

Brubulan Pesucen Spring ranged between 24.97 $dm^3 \cdot s^{-1}$ in December and 145.53 $dm^3 \cdot s^{-1}$ in March (average = 62.83 $dm^3 \cdot s^{-1}$). Each of their monthly hydrographs was gently sloping with an increase in the late rainy season. In response to a rainfall event, Sumbersemen Spring had a relatively prolonged increase in discharge, i.e., in May. As for Brubulan Tahunan and Brubulan Pesucen Springs, their discharge increased in March. Overall, the spring discharges responded slowly to water recharge from rainfall.

Aside from groundwater recharge and drainage, the dynamics of spring discharge are related to the geometry and physical properties of aquifers [MALIK 2015]. Karst spring classification requires data representing the variation of karst spring discharge, which can be acquired from measurements in at least one period of the hydrological cycle. This classification is useful for identifying the stability of spring discharge, which is essential for estimating regional hydrogeological processes and the hydraulics of aquifer and deciding proper spring management [MALIK 2015]. The characteristics of the three major springs in the study area based on average discharge, standard deviation (*SD*), variability index (I_v), variability (*V*), and spring coefficient of variation parameters (*SCVP*) are described in Table 2.

Based on the average values of discharge [MEINZER 1923b acc. to MALIK 2015], Sumbersemen Spring had magnitude 2, Brubulan Tahunan Spring had magnitude 3, and Brubulan Pesucen Spring had magnitude 4. Meanwhile, referring to the Slovak Hydrometeorological Institute [MALIK 2015], the I_v parameter categorized Brubulan Tahunan Spring (I_v = 1.57) and Sumbersemen Spring (I_v = 1.45) into stable springs and Brubulan Pesucen Spring (I_v = 5.83) into an unstable one. As proposed in by MEINZER [1923b] acc. to MALIK [2015], the discharge variabilities defined Brubulan Tahunan Spring (*V* = 46%) and Sumbersemen Spring (*V* = 37%) as fairly constant but Brubulan Pesucen Spring (*V* = 184%) as variable. Based on the *SCVP* [FLORA 2004; SPRINGER *et al.* 2004], Brubulan Tahunan Spring (*SCVP* = 18.58) and Sumbersemen Spring (*SCVP* = 11.46) had low variations, while Brubulan Pesucen Spring (*SCVP* = 63) had a moderate variation.

VARIATION OF THE PHYSICAL AND CHEMICAL SPRING PROPERTIES

The measured physical parameter was temperature, while the chemical parameters included *EC*, pH, major ions, P_{CO2}, and SI_C. The analysis results of these physicochemical properties are summarized in Table 3. They illustrate the level of interaction between $CaCO_3$-H_2O-CO_2 in the karstification process, starting from rainwater falling to surface that initiates mass transport and reactions between the three components [BOGLI 1980].

Based on temperature, Sendang Sayuran Spring had the lowest average (26.59°C), whereas Brubulan Tahunan Spring had the highest average (27.91°C). Brubulan Pesucen and Sumbersemen Springs had a similar temperature, namely 27.01°C and 27.03°C. From January 2018 to

Table 2. The range, average, standard deviation, and characteristics of discharge variations at four major springs in the study area

Spring	*Q* ($dm^3 \cdot s^{-1}$)		*SD*	I_v	*V*	*SCVP*
	range	mean				
Brubulan Tahunan	127.42–199.82	157.71	29.30	1.57	0.46	18.58
Sumbersemen	1 322.22–1 916.67	1 592.33	182.54	1.45	0.37	11.46
Brubulan Pesucen	24.97–145.53	65.58	41.32	5.83	1.84	63.00

Explanations: *Q* = discharge; *SD* = deviation standard; I_v = variability index; *V* = variability; *SCVP* = spring coefficient of variation parameter.
Source: own study.

Table 3. The minimum, maximum, and mean values of the physico-chemical properties of the four major springs in the study area

Parameter	Brubulan Tahunan			Sumbersemen			Brubulan Pesucen			Sendang Sayuran		
	min.	max.	mean	min.	max.	mean	min.	max.	mean	min.	max.	mean
T (˚C)	27.50	28.30	27.91	26.60	27.90	27.01	26.20	27.30	27.03	26.10	27.30	26.59
EC (μS·cm^{-1})	511.00	519.00	513.45	526.00	537.00	529.91	518.00	565.00	541.91	582.00	596.00	587.55
pH	6.82	7.05	6.93	6.90	7.08	6.98	6.86	7.09	6.97	6.82	7.12	6.99
Ca^{2+} (mg·dm^{-3})	93.87	104.83	96.83	87.06	97.92	90.34	94.16	101.95	97.17	120.56	128.35	125.90
Mg^{2+} (mg·dm^{-3})	10.31	12.05	10.95	17.83	21.38	19.12	11.04	21.61	15.88	5.90	10.02	6.90
Na^{+} (mg·dm^{-3})	2.89	4.94	3.37	2.52	4.60	3.07	2.60	6.88	3.28	3.10	5.60	3.50
K^{+} (mg·dm^{-3})	0.80	1.50	1.07	1.25	1.66	1.44	0.79	1.41	1.25	0.48	1.57	0.69
HCO_3^{-} (mg·dm^{-3})	320.36	353.92	328.12	344.76	366.12	351.70	305.10	356.97	332.56	384.43	408.83	394.41
Cl^{-} (mg·dm^{-3})	5.84	11.37	8.89	6.42	9.53	7.91	7.00	9.53	8.37	5.88	9.34	7.54
SO_4^{2-} (mg·dm^{-3})	1.80	8.68	5.05	0.20	8.00	4.87	1.00	8.90	5.42	5.00	20.66	9.37
NO_3^{-} (mg·dm^{-3})	14.95	25.60	22.23	6.90	18.80	15.14	21.30	38.40	31.66	3.20	18.60	10.17
Mg^{2+}:Ca^{2+}	0.16	0.21	0.19	0.31	0.40	0.35	0.18	0.37	0.27	0.08	0.13	0.09
SI_C	–0.12	0.18	–0.01	–0.06	0.19	0.03	–0.10	0.16	0.03	0.05	0.35	0.21
P_{CO2} (%)	3.16	4.90	3.89	3.02	4.27	3.70	2.63	4.27	3.53	2.95	5.75	4.05

Explanations: T = temperature; pH = water acidity measurement; EC = electric conductivity; SI_C = saturation index of $CaCO_3$; P_{CO2} = partial pressure of CO_2.
Source: own study.

January 2019, the pH of the four springs formed a comparable pattern. When the spring discharge began to increase in February and reached its peak in April, the pH decreased due to dilution by rainwater in the recharge area. The pH values later increased from May to January, following a reduction in spring discharge (Fig. 3). A similar pattern of pH fluctuation reflects a similarity in groundwater origin and aquifer [MUSTAFA *et al.* 2015].

Furthermore, the EC value of the four springs did not differ significantly. Sendang Sayuran Spring had the highest EC (average = 588 μS·cm^{-1}), while the EC of the other three springs varied from 513 to 542 μS·cm^{-1}. Except for Brubulan Pesucen Spring with its fluctuating EC, the EC of the other three springs was relatively similar and stable during the measurement period. The EC in Brubulan Pesucen Spring increased as the spring discharge recessed (Fig. 4).

The concentration of Ca^{2+} in the four springs varied with a similar pattern and was relatively stable throughout the observation period (Fig. 5). Sendang Sayuran Spring had the highest and most stable Ca^{2+} concentration. Brubulan Tahunan, Sumbersemen, and Brubulan Pesucen Springs had similar composition and fluctuation of Ca^{2+}. The highest Mg^{2+} variation was detected at Brubulan Pesucen Spring, in which Mg^{2+} concentration increased as the spring discharge decreased.

Meanwhile, in the other three springs, the Mg^{2+} concentration had a similar pattern of variation and was relatively stable (Fig. 6). At Brubulan Pesucen Spring, Mg^{2+} showed a declining trend in January–April, which later increased in May–January. The highest HCO_3^{-} concentration was found at Sendang Sayuran Spring, and it was relatively more stable than at the other springs (Fig. 7). Brubulan Tahunan and Brubulan Pesucen Springs had similar HCO_3^{-} concentration, while Sumbersemen Spring had the lowest concentration. Although the patterns of HCO_3^{-} fluctuation at these four springs were similar and relatively stable, the HCO_3^{-} concentration at Brubulan Pesucen Spring slightly varied. In other words, within this spring, the dilution process by rainwater is visible as the form of H_2O-CO_2 reaction.

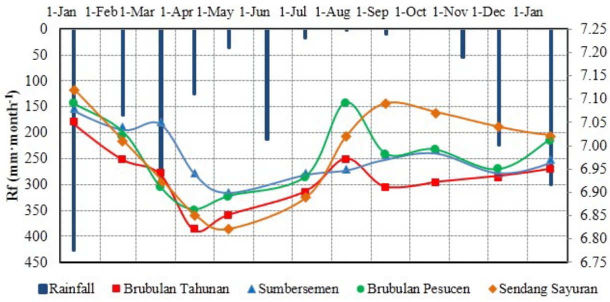

Fig. 3. Monthly rainfall and pH variations at four major springs in the study area; source: own study

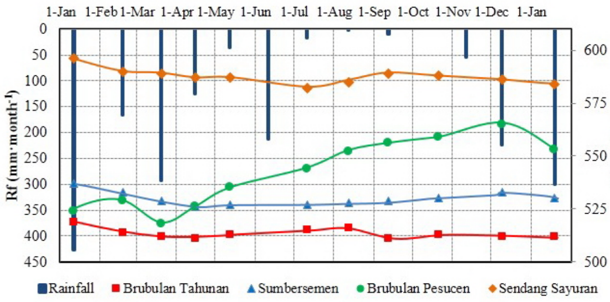

Fig. 4. Monthly rainfall and electrical conductivity (EC) variations at four major springs in the study area; source: own study

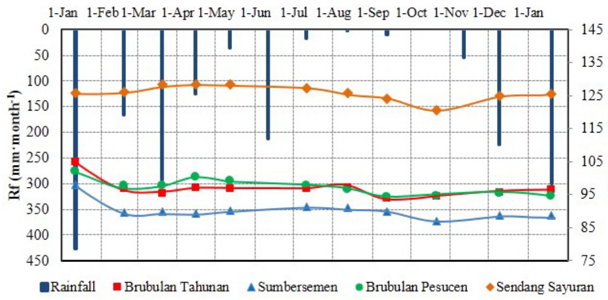

Fig. 5. Monthly rainfall and calcium ion Ca^{2+} concentration variations at four major springs in the study area; source: own study

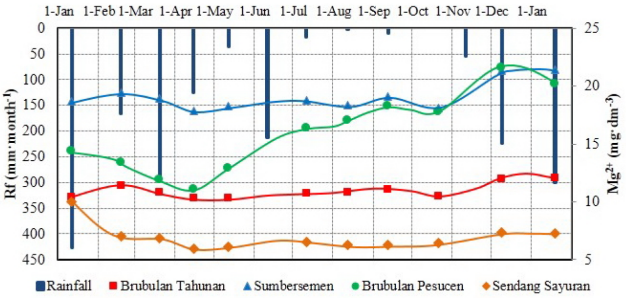

Fig. 6. Monthly rainfall and magnesium ion Mg^{2+} concentration variations at four major springs in the study area; source: own study

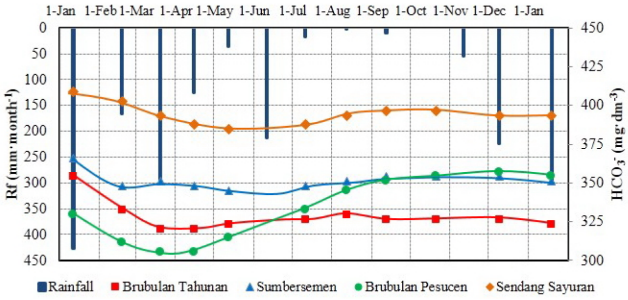

Fig. 7. Monthly rainfall and bicarbonate ion (hydrogencarbonate ion) HCO_3^- concentration variations at four major springs in the study area; source: own study

The variation of P_{CO2} at the four springs created a similar pattern, i.e., an increase in the middle of the rainy season and a peak at the end of the rainy season in May–June (Fig. 8). From the dry season to the early rainy season, P_{CO2} tended to be stable with relatively low values. P_{CO2} at each spring increased as the spring discharge rose, and vice versa. The SI_C is inversely proportional to P_{CO2}, or in other words, a declining trend of SI_C is followed by an increase in spring discharge and vice versa (Fig. 9). Therefore, the SI_C at the four springs tended to have a similar pattern with P_{CO2}. A higher SI_C signifies an increase in CO_2 diffusion in the soil zone [DELBART *et al.* 2014], and when rainwater infiltrates, it is likely to increase spring discharge and P_{CO2} and reduce SI_C. This relationship is not directly apparent in the early rainy season but instead in the middle of the season, as seen in relatively long delay time, i.e., three to four months.

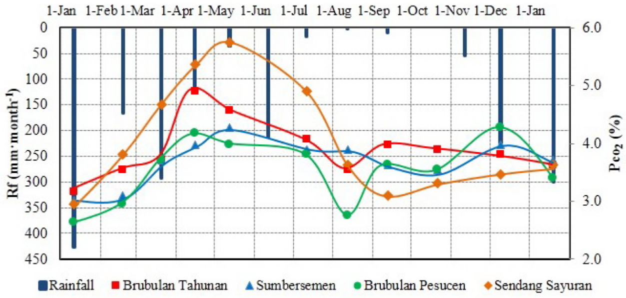

Fig. 8. Monthly rainfall and partial pressure of CO_2 P_{CO2} variation at four major springs in the study area; source: own study

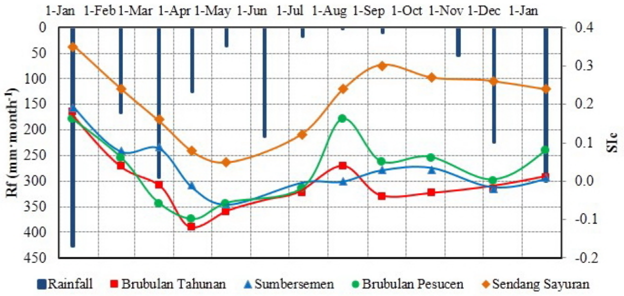

Fig. 9. Graph monthly rainfall and saturation of calcite SI_C variation at four major springs in the study area; source: own study

BIVARIATE CORRELATION OF HYDROCHEMICAL PARAMETERS

The relationship between *EC*, representing the total dissolved ions in water, and the major ions dissolved in karst aquifers, i.e., Ca^{2+}, Mg^{2+}, and HCO_3^-, illustrate the strength of the water–rock interaction in karst aquifer system [KARIMI *et al.* 2005]. Scatter plots connecting *EC* and HCO_3^- proved a positive relationship. The determination (R^2) of both parameters at Brubulan Pesucen Spring was 0.855 (correlation, R = 0.925), Brubulan Tahunan Spring was 0.735 (R = 0.857), Sumbersemen Spring was 0.614 (R = 0.783), and Sendang Sayuran Spring was 0.608 (R = 0.780) – Figure 10a. Strong to very strong positive

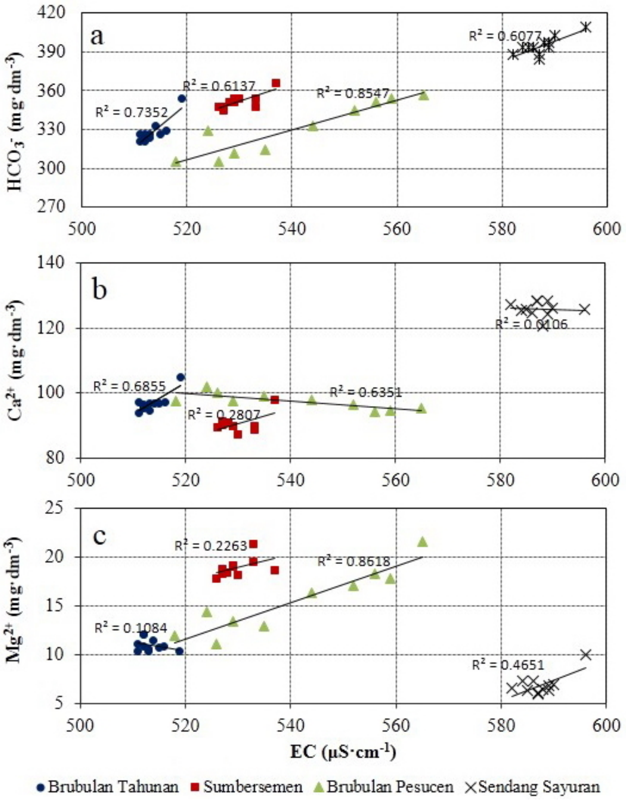

Fig. 10. Correlation of investigated ions concentration and electrical conductivity (*EC*) at four major springs in the study area; a) bicarbonate ion (hydrogencarbonate ion) HCO_3^-, b) calcium ion Ca^{2+}, c) magnesium ion Mg^{2+}; source: own study

correlations between *EC* and HCO_3^- indicate that *EC* is mainly affected by the dissolution of HCO_3^- as a result of CO_2-H_2O-$CaCO_3$ (gas–water–rock interaction). Meanwhile, scatter plots connecting *EC* and Ca^{2+} revealed a diverse relationship. The determination (R^2) of these two parameters at Brubulan Tahunan Spring was 0.686 (R = 0.828), Sumbersemen Spring was 0.281 (R = 0.53), Brubulan Pesucen Spring was 0.635 (R = –0.797), and Sendang Sayuran Spring was 0.011 (R = –0.105) – Figure 10b. Scatter plots of *EC* and Mg^{2+} also showed varying levels of correlation. The determination (R^2) of the two parameters at Brubulan Tahunan Spring was 0.108 (R = –0.329), Sumbersemen Spring was 0.226 (R = 0.475), Brubulan Pesucen Spring was 0.862 (R = 0.928), and Sendang Sayuran Spring was 0.465 (R = 0.682) – Figure 10c.

The positive correlation between *EC* and Ca^{2+} at Brubulan Tahunan Spring shows that the dissolution process, as a result of H_2O–$CaCO_3$ interaction, still occurs due to dilution by infiltrated rainwater. The moderate positive correlation between *EC* and Ca^{2+} and Mg^{2+} at Sumbersemen Spring indicates that infiltration enables dilution by rainwater that affects the dissolution of Ca^{2+} and Mg^{2+}. At Sendang Sayuran Spring, there is no correlation between *EC* and Ca^{2+} but a strong positive correlation between *EC* and Mg^{2+}. It means that although dilution by rainwater affects the dissolution of Mg^{2+}, the opposite is true for Ca^{2+} because Sendang Sayuran Spring was saturated with $CaCO_3$ throughout the research period. A strong negative correlation between *EC* and Ca^{2+} and a strong positive correlation between *EC* and Mg^{2+} were detected at Brubulan Pesucen Spring. These findings indicate that the process of Mg^{2+} enrichment is followed by a reduction of Ca^{2+}. This process is apparent from the Mg^{2+}:Ca^{2+} molar ratio at Brubulan Pesucen Spring, which increases as the spring discharge decreases. At the end of the rainy season, the Mg^{2+}:Ca^{2+} molar ratio of this spring was similar to that of Sumbersemen Spring (Fig. 11).

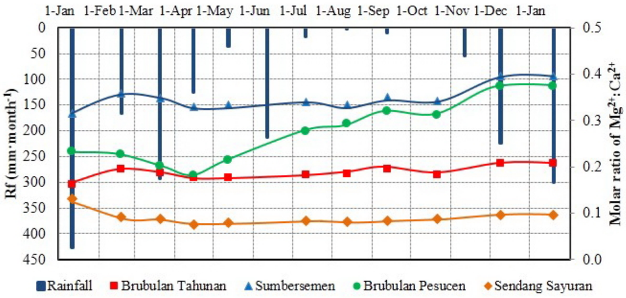

Fig. 11. Monthly rainfall and Mg^{2+}:Ca^{2+} ratio variations at four major springs in the study area; source: own study

The presence of Mg^{2+} in water samples can be used as an indicator of the residence time of groundwater in aquifers [HISCOCK, BENSE 2014]. A higher Mg^{2+}: Ca^{2+} ratio signifies a longer residence time because Mg^{2+} is released from groundwater at a slower rate than Ca^{2+} [BATIOT *et al.* 2003]. Based on this ratio, Sumbersemen Spring has a relatively distant source and prolonged contact because the groundwater recharge area is at Gutak Hills. Meanwhile, in dry seasons, the source of groundwater at Sumbersemen Spring can compensate for the low water level at Brubulan Pesucen Spring.

DISCUSSION

GROUNDWATER FLOW CHARACTERISTICS

The karst groundwater flow character is related to the groundwater movement from the recharge to the discharge area, such as springs, and it can be identified from the hydrograph characteristics of springs [SMART, HOBBES 1986]. Monthly discharge measurement at three major springs from January 2018 to January 2019 produced discharge hydrographs, as seen in Figure 12. When compared with the hypothetical model of karst groundwater recharge, storage, and flow system proposed by SMART and HOBBES [1986], the discharge measurement results showed a combination of conduit and diffusion flow, with diffusion as the more substantial flow component, especially at Brubulan Tahunan and Sumbersemen Springs. This figure also indicates that the groundwater recharge system is dominated by a dispersed rather than concentrated mechanism. A slightly different result was found at Brubulan Pesucen Spring. In this spring, the conduit network is more developed than the other two springs. Therefore, the generated hydrograph has a steeper increasing limb.

Karst aquifers can also be characterized by analyzing the *FDC* [FLOREA, VACHER 2006; KATSANOU *et al.* 2015]. The *FDC* of a simple karst system has a straight trend line with a particular slope, whereas that of a complex karst system shows a variation in the trend line that is attributable to discharge losses toward an adjacent unit or, conversely, a supplementary recharge [SOULIOS 1991]. Observation on the *FDC*s showed that the three springs had a complex system, as apparent from the steep and very steep curves of Brubulan Tahunan and Sumbersemen Springs and the gently sloping curve of Brubulan Pesucen (Fig. 13).

Based on the characters of the generated *FDC*s, the three springs above show a similarity, that is, a single outlet system [KATSANOU *et al.* 2015]. The curves of Sumbersemen and Brubulan Tahunan Springs have slope breaks, respectively at >1370 $dm^3 \cdot s^{-1}$ and >140 $dm^3 \cdot s^{-1}$, meaning that both springs emerge from rapid and stable flow system and supplementary recharge or, in other terms, a storage with large capacity and low variability [FLOREA, VACHER 2006]. The *FDC* of Brubulan Pesucen changes to a gently sloping curve at the discharge of >33 $dm^3 \cdot s^{-1}$, which demonstrates rapid drainage with relatively low supplementary recharge through the conduits. These curve features confirm that the conduit system of Brubulan Pesucen is more developed than Sumbersemen and Brubulan Tahunan. The same case is observable from the outflows of Brubulan Pesucen that show horizontal cavities and Sumbersemen and Brubulan Tahunan, which suggest artesian springs that form "spring pools".

In addition to hydrographs and *FDC*, the characteristics of groundwater flow in karst springs can be identified from the frequency of changes in the chemical properties and hydraulics of springs, whether as conduit or diffusion

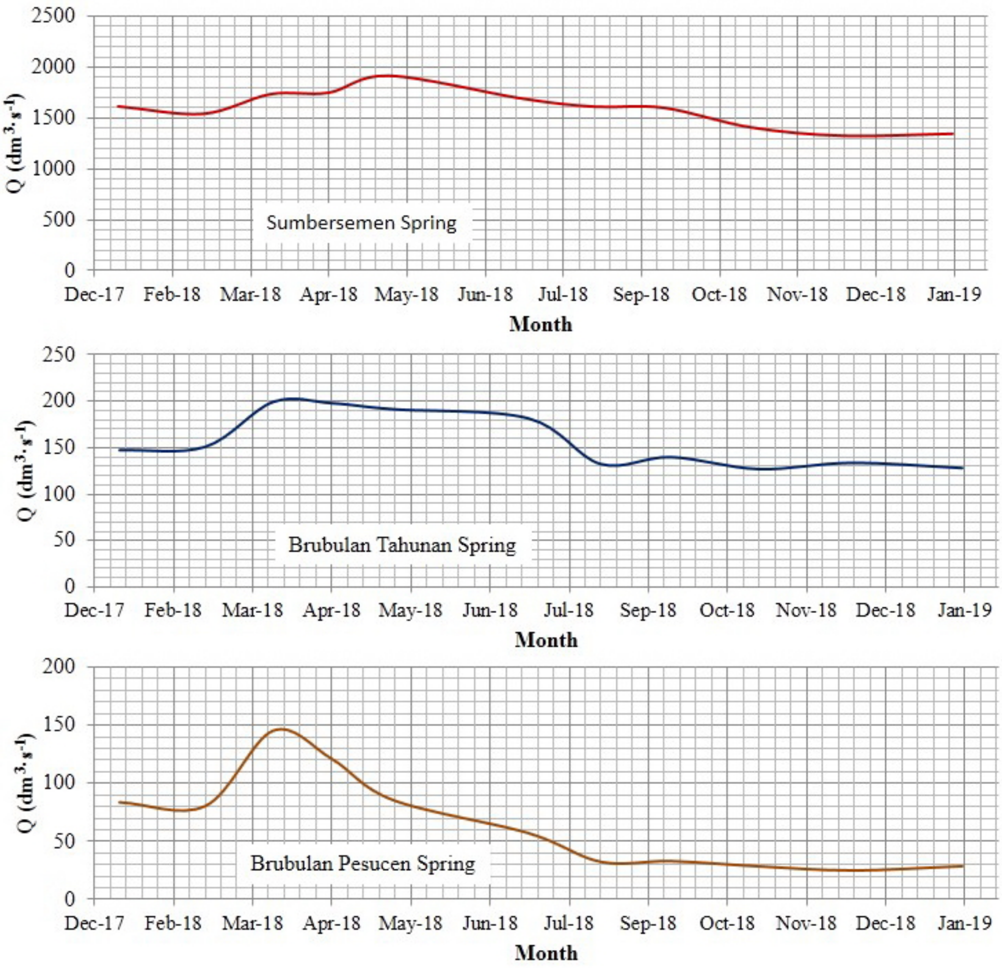

Fig. 12. Monthly discharge hydrographs at three major springs in the study area; source: own study

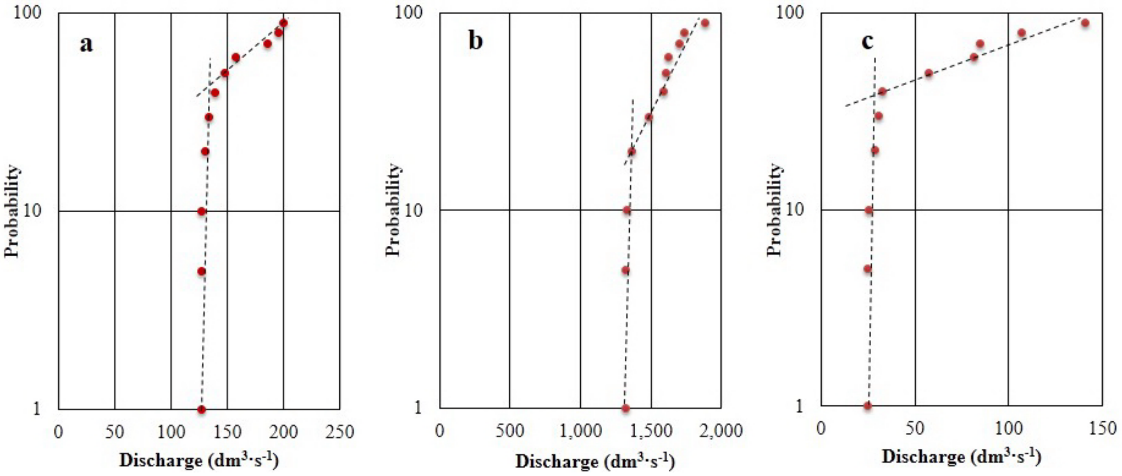

Fig. 13. Flow duration curves (FDC) of three major springs in the study area: a) Brubulan Tahunan, b) Sumbersemen, c) Brubulan Pesucen; source: own study

system [SMART, HOBBES 1986]. This factor is represented by the coefficient of variation in the physico-chemical water parameters, such as *EC*, temperature, hydrochemical, and spring discharge [BAENA *et al.* 2009]. The coefficient of variation (*CV*) is the standard deviation divided by the average value multiplied by 100 [EFTIMI 2005]. SHUSTER and WHITE [1971], JACOBSON and LANGMUIR [1974] and EFTIMI [2005] correlate the properties of groundwater flow at several springs with variations in discharge and physico-chemical properties of groundwater, such as temperature, *EC*, Ca^{2+}, Mg^{2+}, and HCO_3^-. They claim that a conduit spring has a relatively high coefficient of variation, while a diffusion spring has a low one (Tab. 4).

Table 4. The comparison of coefficients of variation of several physico-chemical water parameters from some previous studies

Spring typology	Coefficient of variation *CV*							Source
	T	*Q*	*EC*	pH	Ca^{2+}	Mg^{2+}	HCO_3^-	
Rock Spring conduit type	26.9	175	23	1.7	25.5	27.6	28.6	JACOBSON and LANGMUIR [1974]
Iskert Spring conduit type	10.3	94.2	–	1.6	11.7	30	14.2	JACOBSON and LANGMUIR [1974]
Conduit type	26.9	–	10–20	–	–	–	–	SHUSTER and WHITE [1971]; JACOBSON and LANGMUIR [1974]
Thomson Spring diffusive type	1.4	26.3	6.2	0.6	6.4	2	2.7	JACOBSON and LANGMUIR [1974]
Blue Eye diffusive type	0.6	–	5	–	–	–	–	EFTIMI [2005]
Pellumbas diffusive type	4.2	–	7.2	–	12.6	23.5	–	EFTIMI [2005]
Diffusive type	1.4	–	< 10	–	–	–	–	SHUSTER and WHITE [1971]; JACOBSON and LANGMUIR [1974]

Explanations: *T* = temperature; *Q* = discharge; *EC* = electric conductivity; pH = measurement of water acidity.
Source: own elaboration based on literature data.

Table 5. The coefficients of variation (*CV*) of the physico-chemical properties of four major springs in the study area

Spring	*CV*						
	T	*Q*	*EC*	pH	Ca^{2+}	Mg^{2+}	HCO_3^-
Brubulan Tahunan	1.04	18.58	0.47	0.89	2.95	5.69	2.83
Sumbersemen	1.55	11.46	0.62	0.80	3.06	6.26	1.60
Brubulan Pesucen	1.19	63.00	2.98	1.09	2.61	21.75	6.24
Sendang Sayuran	1.69	–	0.62	1.44	1.82	16.41	1.77

Explanations as in Table 4.
Source: own study.

The coefficients of variation of the physico-chemical spring properties in the study area are presented in Table 5. The variations in the amount of discharge and the physico-chemical properties of springs were generally low. A relatively high variation was found in the physico-chemical parameters of Brubulan Pesucen Spring. Compared with the other springs, Brubulan Pesucen Spring had higher variations in discharge, *EC*, and Mg^{2+} and HCO_3^- compositions. A relatively high variation of Mg^{2+} composition was also detected at Sendang Sayuran Spring.

The coefficients of variation defined the four springs in the study area as diffusion karst springs, as suggested by SHUSTER and WHITE [1971], JACOBSON and LANGMUIR [1974] and EFTIMI [2005]. In a diffusion system, groundwater flows through micro-fractures in karst rocks and has a less fluctuating discharge [WHITE 1988]. Although the study area has many geological structures, the variations in discharge and hydrochemical properties indicate that the groundwater is stored in a densely fractured rock (micro-fracture block). This finding supports RAEISI, KARAMI [1997] and OZYURT, BAYARI [2008], which explain that groundwater stored in a matrix produces a diffusion flow with long residence time.

Relatively prolonged responses of spring discharge and hydrochemical characteristics to rainfall (up to 3–4 months) show that a large number of ponors and sinkholes in the study area are not yet connected as a conduit network system described by SMART and HOBBES [1996]. GILLIESON [1996] state that this aquifer mostly has a mixed type between diffuse and conduit or, in other words, water comes from conduit, fracture, and matrix or pore systems. This conduit has a single outlet system, confirming the *FDC* analysis results, and receives groundwater flow from storage composed of dense fractures and porous media. Geological structures, namely faults and folds, primarily control its development. GOLDSCHEIDER [2005] affirm that faults and folds are the main factors in speleogenesis and the development of subsurface drainage in the karst system. The existence of dense fractures and porous media indicates that the groundwater is stored in various aquifers, i.e., karst (limestone) and non-karst aquifers (composed of stratigraphy with alternating sandstone, limestone, and quartz sandstone).

A mixed-flow system indicates that the mechanism of groundwater infiltration in the recharge area also has a mixed type, which according to FORD and WILLIAMS [2007] and GOLDSCHEIDER [2015], is a combination of direct recharges from karst rocks (autogenic recharge) and non-karst rocks (allogenic recharge). This type of flow is possible because at above 155 m a.s.l. (the lowest elevation of spring with the most significant discharge, Sumbersemen), the study area is composed of complex formations, namely the limestone of the Tawun Formation, the quartz sandstone of the Ngrayong Formation, the limestone of the Bulu Formation, the sandstone with limestone intercalations of the Wonocolo Formation, and the limestone of the Paciran Formation [KADAR, SUDIJONO 1993]. These formations cover a wide area from the Watuputih Hills to the western hills with the west-east direction, which is controlled by a syncline-anticline system, i.e., the Ngiono-Pakel anticline and Bulu syncline [KADAR, SUDIJONO 1993] – Figure 14. In other words, the groundwater recharge system has a combination of autogenic and allogenic recharge; both of which are controlled by anticlinorium hills. The autogenic recharge comes from the limestones of the Tawun, Bulu, and Paciran Formations, while the allogenic recharge is provided by the quartz sandstone of the Ngrayong Formation and the sandstone with limestone intercalations in the Wonocolo Formation.

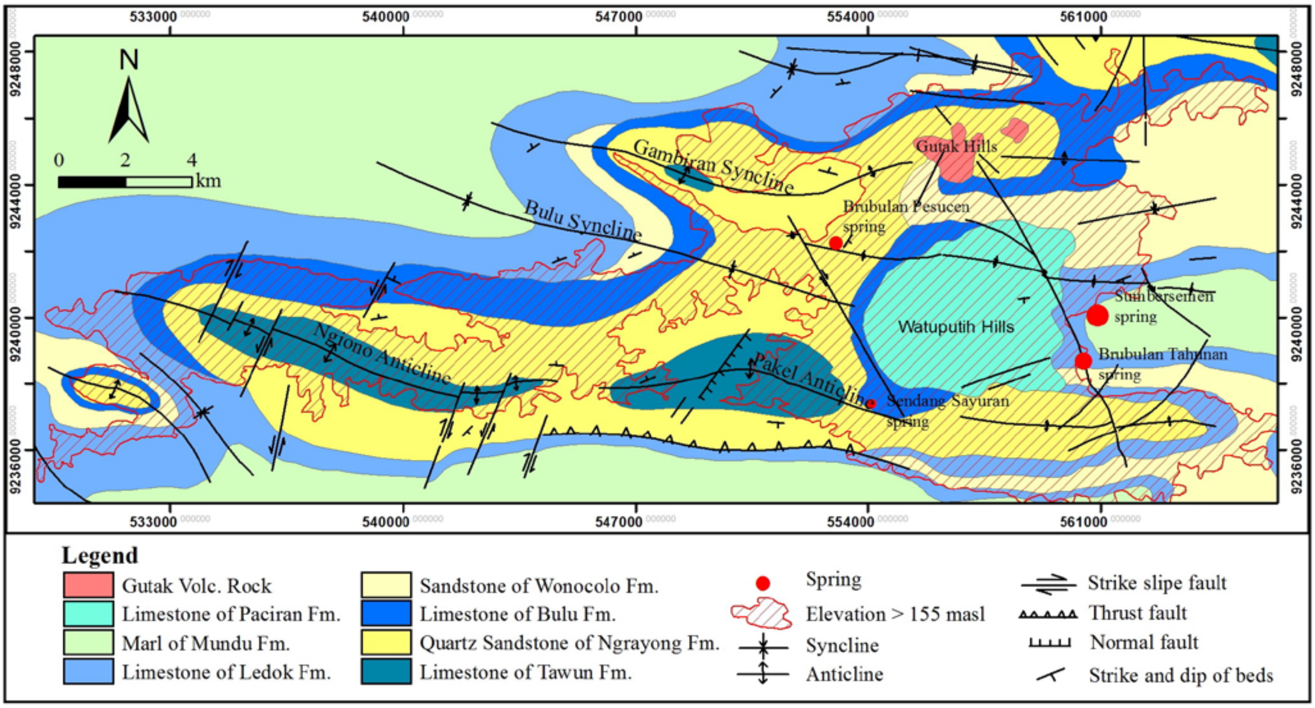

Fig. 14. The zone above 155 m a.s.l. showing the diverse rock formations and geological structures of the study area based on a 1:100,000 geological map; source: own elaboration

The autogenic recharge system involves two mechanisms, namely dispersed and concentrated. The recharge mechanism by diffusion is the product of the relatively thick epikarst layer of the limestone in the study area, while concentrated recharge comprises ponors and sinkholes developing in the limestones of the Bulu and Tawun Formations. The observation in the field proved that the karstification process in the limestone of the Paciran Formation is undeveloped, as marked by the rare formations of ponors and sinkholes. Besides, the surface drainage patterns follows the non-karst rocks. Meanwhile, allogenic recharge system occurs by dispersion mechanism, and no surface flow system enters the karst aquifer in a concentrated manner.

HYDROGEOLOGICAL SYSTEM

The karst hydrogeological system in the study area is formed in the anticlinorium zone of Rembang, which has unique groundwater recharge, flow, and discharge systems in the form of springs. The recharge system is a mixing of autogenic (derived from limestone) and allogenic (derived from sandstones). The groundwater drainage system also shows a fast flow due to the influence of geological structures, but it also has a slower-release storage system. This combination produces a spring system with large and stable discharge. According to OZYURT and BAYARI [2008], this phenomenon occurs because high hydraulic conductivity zones are filled quickly by the surrounding aquifer storage that has smaller hydraulic conductivity (densely fractured rocks or matrices). Therefore, the springs in the study area are different from karst springs in general. Springs with a diffusion system have relatively small discharges with low variation and less fluctuating hydrochemical compositions, whereas the ones with a conduit system have relatively large discharge with high variation and widely fluctuating hydrochemical compositions [RAEISI, KARAMI 1997].

Conceptually, the hydrogeological system in the study area is depicted in Figure 15. The subsurface conditions were reconstructed based on a geophysical survey using both gravity and resistivity methods [GAI 2017]. The geological cross-section shows that a fault system also develops in the middle of the study area and consists of faults cutting through the limestone of the Tawun Formation, the quartz sandstone of the Ngrayong Formation, and the limestone of the Bulu Formation. These faults are not visible on the surface because they are covered by the limestone of the Paciran Formation that forms the Watuputih Karst Hills. Due to these geological structures, the groundwater from the sandstone aquifer of the Ngrayong mixes with the limestone aquifer system before emerging on the surface as a spring.

Aside from the gently sloping hydrograph, the P_{CO2} and SI_C chemographs also demonstrate the slow response of groundwater flow to rainwater (recharge). Groundwater recharge by rainwater infiltration to the aquifer layer triggers CO_2-H_2O-$CaCO_3$ interaction and causes an increase in P_{CO2} and a decrease in SI_C [ADJI *et al.* 2016; DELBART *et al.* 2014]. Elevated P_{CO2} and reduced SI_C occurred as the spring discharge in the middle of the rainy season increased, which was evident from a relatively prolonged delay time from three to four months (Figs. 8, 9). Moreover, the water sample analysis also proved that the springs were undersaturated with $CaCO_3$ in the early dry season. An exception applies to Sendang Sayuran Spring, which

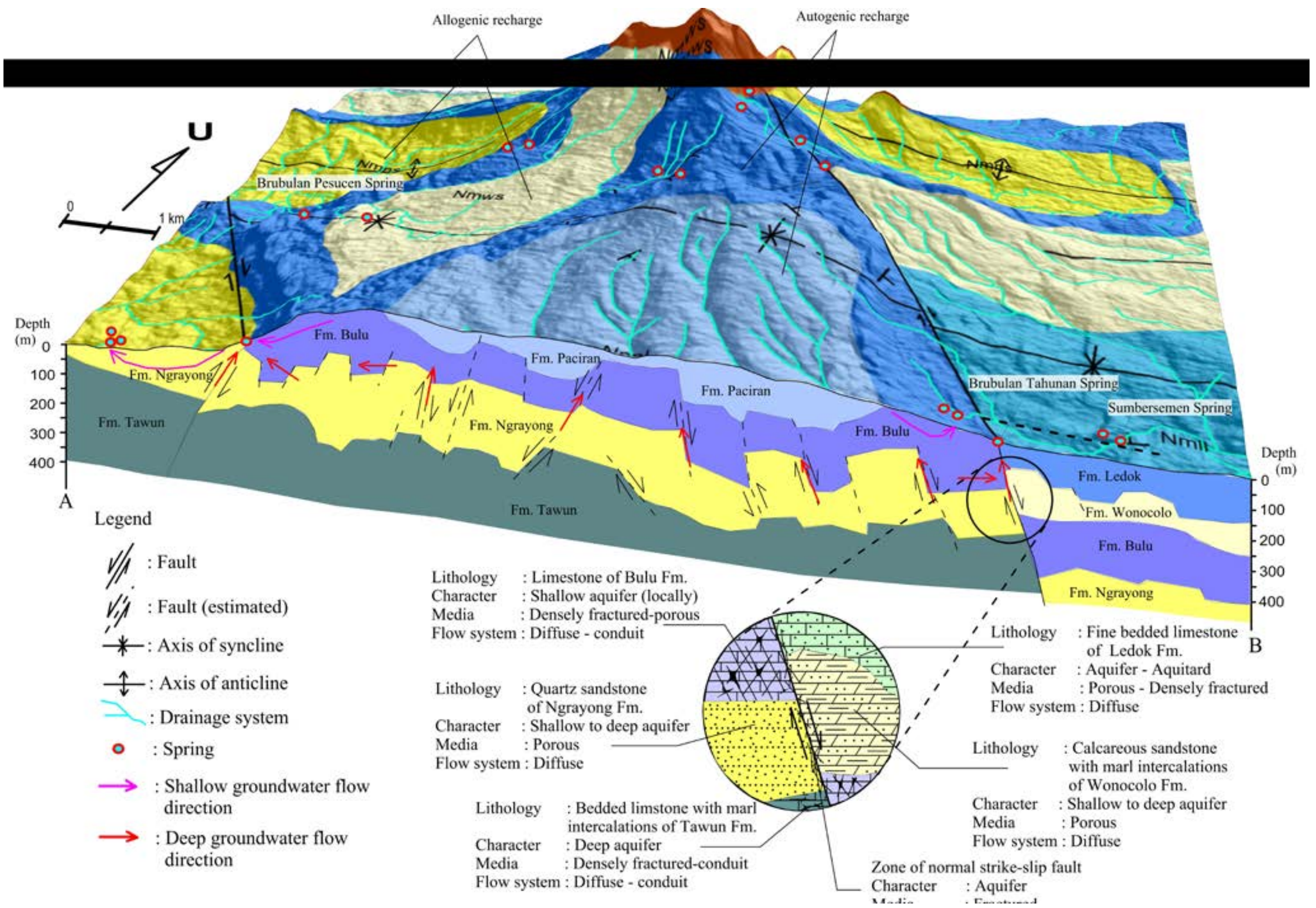

Fig. 15. The conceptual model of the hydrogeological system in the study area; source: own elaboration

was relatively saturated throughout the measurement period. Brubulan Tahunan Spring, located in a normal fault, experienced the longest $CaCO_3$-unsaturated condition ($SI_C < 0$), from March to December (Fig. 9), which is potentially caused by the dilution effects of the local groundwater recharge near the fault zone and the Watuputih Hills above it. The temperature data also support this finding. The temperature at Brubulan Tahunan Spring (27.91°C) was the highest (other springs T = 26.59–27.03°C), which indicates a slight influence from the atmospheric conditions.

Based on variations in the amount of discharge and the hydrochemical profiles, the four springs have the same hydrogeological system, including the groundwater recharge, flow, and discharge. However, each spring still has a unique variation in its hydrochemical characteristics. Sumbersemen Spring, located on a syncline and a fault, has the most abundant discharge and is the most stable spring. It means that the hydraulic gradient and syncline system control the magnitude and stability of the spring discharge, considering that it is located on a synclinal limb and at the lowest elevation (155 m a.s.l.). Brubulan Tahunan Spring, which is controlled by a normal fault, has the second-largest discharge and is also a relatively stable spring. It is located at the same elevation as Sumbersemen Spring. Brubulan Pesucen Spring emerges at the same syncline system as Sumbersemen Spring, but the former has a much smaller and more variable discharge than the latter. This finding indicates an association with the elevation of Brubulan Pesucen Spring, which is higher than Sumbersemen Spring, i.e., 237 m a.s.l. In other words, the geological structure and hydraulic gradient formed in the groundwater recharge and discharge area significantly determine the amount of spring discharge.

Sendang Sayuran Spring is located in a plunging anticline composed of limestone from the Tawun Formation (Nmtl) and at the highest elevation (288 m a.s.l.) among the four springs. It has higher Ca^{2+} and HCO_3^- compositions with lower variation (stable) than the other springs; hence, the highest mineral saturation. It is also composed of the lowest Mg^{2+}, as evident from the Ca-HCO_3 facies throughout the measurement period. This finding indicates that the recharge, drainage, and discharge systems of this spring take place in a relatively homogenous rock, namely the limestone of Tawun Formation (Nmtl) – the oldest rock in the study area. This spring was saturated with $CaCO_3$ throughout the research period or, in other words, the deposition of $CaCO_3$ was dominant, which is proven by the formation of travertine layers around the spring. This condition represents the less developed conduit network in the limestone of the Tawun Formation.

Sumbersemen Spring has the highest Mg^{2+} composition because of the Ca-Mg-HCO_3 groundwater facies throughout the measurement period. It indicates that the source of groundwater at Sumbersemen Spring has a relatively long contact with the aquifer materials and is also related to the groundwater recharge around Gutak Hills. Brubulan Pesucen Spring also has a high Mg^{2+} composition with a wide variation and negative correlation with the spring discharge. This composition was apparent from the change in the hydrochemical facies from Ca-HCO_3 in January–May 2018 to Ca-Mg-HCO_3 in July 2018–January 2019. This process indicates that the groundwater recharge of Sumbersemen Spring can compensate the Brubulan Pesucen Spring discharge in the dry season, which is possible because both springs are located in the same syncline system.

CONCLUSIONS

The spatio-temporal data analysis of the spring discharge and physico-chemical properties (*T*, pH, *EC*, Ca^{2+}, Mg^{2+}, and HCO_3) of the four main springs in the Watuputih Hills have identified the characteristics of the aquifer system. The variability index (I_v), variability (*V*), and spring coefficient of variation parameters (*SCVP*) classify Brubulan Tahunan and Sumbersemen as stable, fairly constant springs with low variations and Brubulan Pesucen as an unstable and variable spring with moderate variations. The findings include a gently sloping hydrograph, low variations in discharge and hydrochemical properties, a relatively long response of discharge and CO_2-H_2O-$CaCO_3$ interaction to rainfall, and slope breaks on the flow duration curve (*FDC*). These indicate that even though faults and folds that are conduit in nature control the springs, there are diffuse storage media in the forms of dense fractures and pores. They also illustrate a less developed interconnected conduit network, although Brubulan Pesucen is relatively more developed than Sumbersemen and Brubulan Tahunan. The dynamics of the discharge and hydrochemical characters prove that, hydrogeologically, the three springs have the same recharge, flow, and discharge systems that involve both karst and non-karst rocks (sandstones). The geological structure and hydraulic gradient formed between the groundwater recharge and discharge areas provide valuable information for spring discharge management. Sumbersemen Spring, situated on a synclinal limb and at the lowest elevation (155 m.s.l.), has the largest and most stable discharge, averagely 1592 $dm^3 \cdot s^{-1}$, while Brubulan Tahunan Spring, located in a normal fault, releases an average discharge of 158 $dm^3 \cdot s^{-1}$. Along with the decrease in spring discharge, Mg^{2+} enrichment in Brubulan Pesucen Spring indicates that the groundwater coming from the same source as Sumbersemen Spring can compensate for the low flow rate in the dry season, especially considering that both springs are located in the same syncline system. Sendang Sayuran Spring, located in a plunging anticline, has the highest Ca^{2+} and HCO_3^- compositions and $CaCO_3$ saturation, as well as a stable character. These findings represent a relatively homogenous aquifer system, namely the limestone of the Tawun Formation (Nmtl). However, this research is a “preliminary study” because the analyzed discharge hydrograph presents a monthly data that cannot explain the characteristics of groundwater flow, such as the proportion of diffusion and conduit flow generated from the karst aquifer storage system, in detail. Further detailed research is required, and it needs to install climatology stations and loggers that measure spring discharge on a daily or hourly basis.

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