

Hydrogeological modelling of geothermal waters in Pamukkale, western Anatolia, Turkey

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The study area, located in the eastern part of the continental rift zone of the Büyük Menderes within the Menderes Massif, western Anatolia, is composed of Paleozoic metamorphic rocks, Mesozoic limestones and Eocene via Pliocene to Quaternary sediments. Paleozoic marbles, quartzites and carbonate schists, Mesozoic limestone, Pliocene sediments and Quaternary alluvium and travertine serve as permeable rocks for the geothermal waters. The geothermal waters in Pamukkale and environs with outlet temperatures of about 35 °C and reservoir temperatures up to 250 °C can be considered as Ca-Mg-SO₄-HCO₃ type waters. The formation of the travertine in Pamukkale is one of the world's wonders, directly connected with decreasing temperatures and CO₂ partial pressures. The formation of travertine deposits depends upon the solubility of CaCO₃ controlled principally by CO₂ partial pressure, temperature and pH values, in which reaction equilibria play an important role. Moreover, the travertine deposits, which show a U-series age of at least 400 ka form one of the important world wonders. The geothermal waters of Pamukkale, with its high sulphate contents up to 650 mg/l and Rn concentrations up to 20 Bq/l, were modelled hydrogeologically from schematical points.

La zone étudiée, localisée dans la partie orientale de la zone de rift continentale du Büyük Menderes, à l'intérieur du massif de Menderes, en Anatolie occidentale, est constituée de roches métamorphiques paléozoïques, de calcaires mésozoïques et de sédiments d'âges éocène, pliocène à quaternaire. Les marbres du Primaire, les quartzites et les calcaires schisteux, les calcaires mésozoïques, les sédiments pliocènes et les alluvions et travertins quaternaires représentent des formations perméables pour les eaux géothermales. Celles-ci à Pamukkale et ses environs, présentant, à la sortie, des températures d'environ 35 °C et des températures de réservoir pouvant atteindre 250 °C, peuvent être considérées comme des eaux de composition contenant les éléments Ca-Mg-SO₄-HCO₃. La formation de travertin à Pamukkale est l'une des merveilles du monde, directement liée aux baisses de température et, en partie, de pression du CO₂. La formation des dépôts de travertin dépend de la solubilité du CaCO₃, contrôlée principalement par, pro parte, la pression de CO₂, la température et les valeurs de pH, solubilité pour laquelle les équilibres de réaction jouent un rôle important. De plus, ces dépôts, d'âge minimum 400 000 ans -datation à l'uranium- constituent aussi une merveille du monde, significative. Les eaux géothermales de Pamukkale avec leur haute concentration de sulfate (jusqu'à 650 mg/l), et de radon (jusqu'à 20 Bq/l) ont fait l'objet d'une modélisation hydrogéologique à partir d'un échantillonnage de points.

El área de estudio, situado en la parte oriental de la zona del rift continental de los Büyük Menderes dentro del Macizo de Menderes, Anatolia occidental, está compuesta de rocas metamórficas paleozoicas, calizas mesozoicas y eoceno a través de pliocenos a sedimentos cuaternarios. Los mármoles paleozoicos, las cuarcitas y los carbonatos esquistos, la piedra caliza mesozoica, los sedimentos del Plioceno, el aluvión cuaternario y el travertino sirven como rocas permeables para las aguas geotérmicas. Las aguas geotérmicas de Pamukkale y sus alrededores con temperaturas de salida de aproximadamente 35 °C y temperaturas de depósito hasta 250 °C pueden considerarse como aguas de tipo Ca-Mg-SO₄-HCO₃. La formación del travertino en Pamukkale es una de las maravillas del mundo, directamente conectado con la disminución de las temperaturas y las presiones parciales del CO₂. La formación de depósitos de travertino depende de la solubilidad del CaCO₃ controlado principalmente por la presión parcial del CO₂, la temperatura y los valores de pH, en los cuales los equilibrios de reacción juegan un papel importante. Por otra parte, los depósitos de travertino, que muestran una edad de la serie U de al menos 400 ka forman una de las maravillas mundiales importantes. Las aguas geotérmicas de Pamukkale, con su alto contenido de sulfatos hasta 650 mg/l y concentraciones de Rn hasta 20 Bq/l, fueron modeladas hidrogeológicamente desde puntos esquemáticos.

In Turkey, geothermal waters are located in large areas in connection with tectonic features and volcanism from the Middle Miocene to recent in age. The high-enthalpy geothermal waters form in the continental rift zones of the Menderes Massif, which suffered compression and later extension tectonics. The study area

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of Pamukkale is located 20 km NW of the provincial capital of Denizli (Figures 1 and 2) which is 360 m above sea level. The geothermal waters of Pamukkale are located in the southern shoulder of Çökelez Mountain, in the travertine platform and within the travertine mass forming an unrivalled example in the world. The area of travertine and the antique ruins of the Hierapolis City form an important centre due to its original natural structures and historical value. The study area of Pamukkale has an area of 44 km² and is a Special Environment Pro-

tection Region with five residential areas: Develi, Karahayit, Pamukkale, Yeniköy and Akköy. The aim of this study is (i) to update geological mapping of the geothermal areas in Pamukkale and environs, (ii) to describe the water-rock interaction by mineralogical, petrographical and geochemical working methods, (iii) to investigate the formation and development of geothermal waters by hydrogeological, hydrogeochemical and isotope geochemical methods and (iv) to create a conceptual hydrogeological model of the Pamukkale geothermal field.

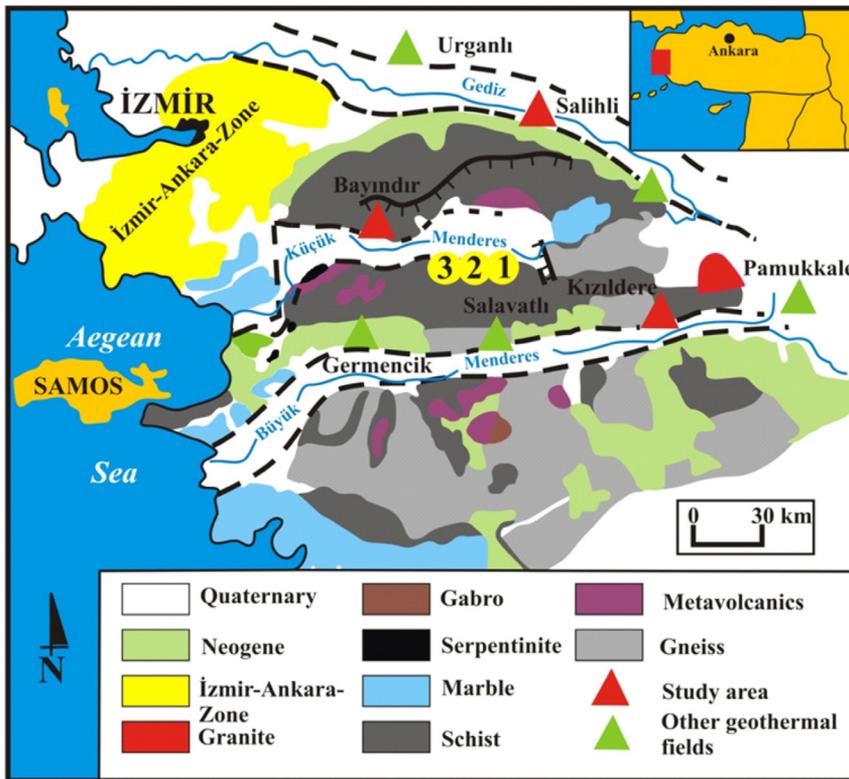


Figure 1: Geological map of the Menderes Massif and the location of the study area of Pamukkale. 1: Mercury deposit of Haliköy, 2: Antimony deposit of Emirli, 3: Arsenopyrite and gold deposit of Küre (modified from Özgür, 1998).

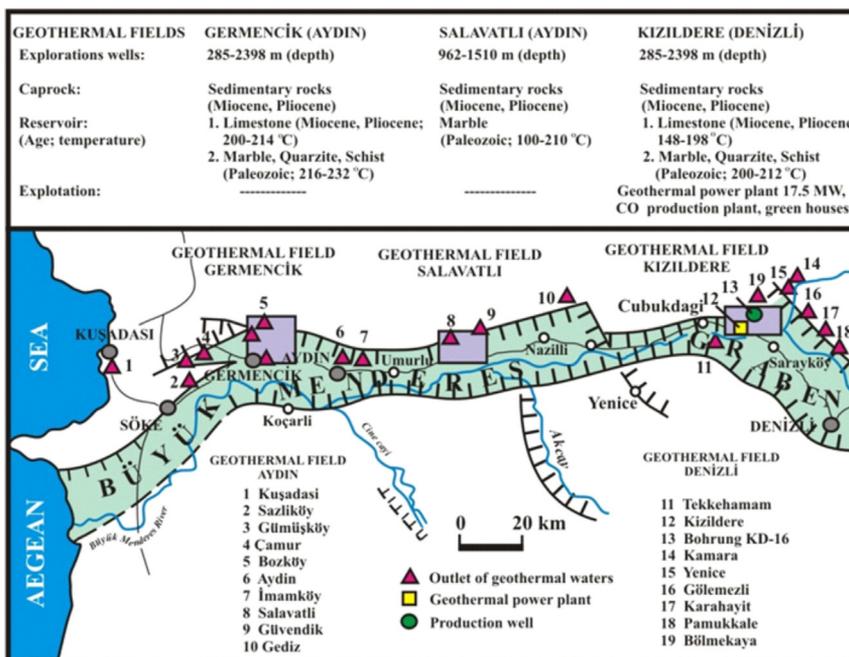


Figure 2: Geothermal waters in the rift zone of the Büyük Menderes and the location of Pamukkale (modified from Özgür, 1998).

Material and methods

In order to understand the hydrogeochemical features of the geothermal waters in Pamukkale and environs, 16 samples

were collected (Kıymaz, 2011; Kutlu, 2015; Uzun, 2017) (Figure 3; Table 1). During this sampling campaign, in-situ measurements such as temperature, pH, Eh, electrical conductivity and alkalinity were taken

(Table 1). The cations of Na^+ , Ca^{2+} , Mg^{2+} , K^+ , Si^{4+} and B^{3+} were analysed by ICP-OES methods, while the analyses of anions such as F^- , SO_4^{2-} , Cl^- and NO_3^- were performed by IC methods. The values of HCO_3^- and CO_3^{2-} have been calculated by the alkalinity measurements in the field. The evaluation of the hydrogeochemical data was carried out using Aquachem 3.7 (Calmbach, 1999).

Results

Geologic setting

Denizli Basin is located in the Aegean Region, where the E-W trending continental rift zone of the Büyük Menderes – limited by active and normal faults – and the NW-SE trending continental rift zone of the Gediz incorporate (Figure 4). In the study area, Paleozoic metamorphic rocks of the Menderes Massif form the basement rocks which are overlain by Mesozoic limestones, Eocene to Pliocene sedimentary rocks and Quaternary alluvium and travertine. The travertine of Pamukkale precipitates on the falling block of the Pamukkale fault, which is located in the eastern part of the basin and constrains the basin in the North (Altunel, 1996). In the areas with intensive fissures of the main fault, intensive precipitations of travertine can be observed. Parallel to oblique fissures were generated in connection with main Pamukkale fault. In the study area, opening fissures were observed, and ridge type travertine has been observed in some of the fissures. With the exception of Pamukkale, the travertine of Denizli can be observed in localities such as Yeniköy, Küçükdereköy, Irlıganlı, Kocabaş, Koyunaliler and Karateke in the eastern direction. The factors affecting the precipitation of travertine are (i) the compositions, saturation indexes and partial CO_2 pressures of geothermal and mineral waters, (ii) the temperatures, flow regimes and flow rates of the geothermal waters and (iii) the temperature of the geothermal waters during flow.

Hydrogeology

In the study area, Paleozoic marbles, Mesozoic limestones, Pliocene sediments and Quaternary alluviums and travertine occur as permeable rocks in general. Paleozoic marbles can be observed between Karahayıt and Pamukkale and in the NE part of the Pamukkale main springs, whereas Mesozoic limestones occur to the north of Pamukkale main springs. Pliocene sediments are found in the environs of Pamukkale main springs and in the upper part of Yenice horst between Pamukkale and Karahayıt.

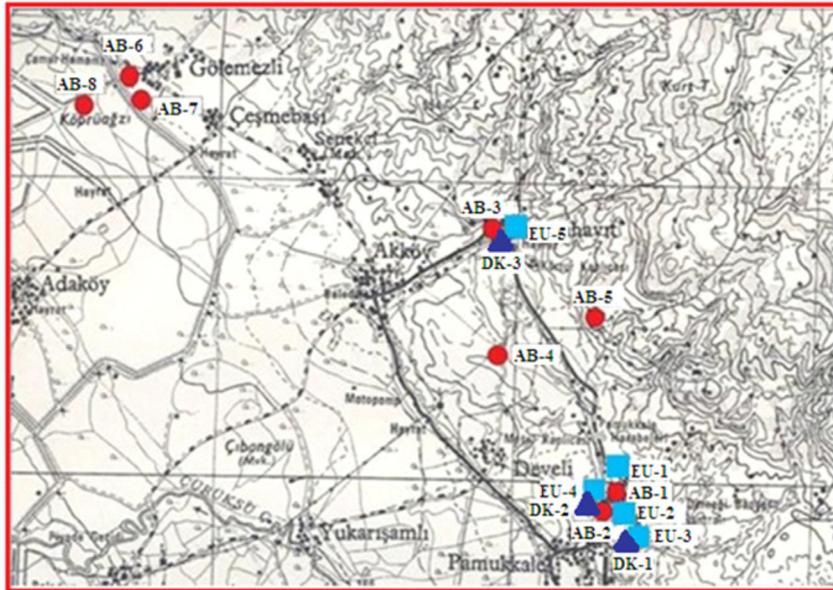


Figure 3: Sample locations of the geothermal field of Pamukkale (modified from Kıymaz, 2011; Kutlu, 2015; Uzun, 2017).

The Kolonkaya and Tosunlar formations on the first shallow reservoir rock form an intercalation of claystones, marls and sandstones and are good cap rocks for the first reservoir rocks. These cap rocks have a thickness of 350– 600 m.

Hydrogeochemistry

The geothermal waters of Pamukkale and environs can be considered as Ca-Mg-(SO₄)-HCO₃ type waters in the Piper diagram (Figure 5). Hydrogeochemically, the geothermal waters in the study area display the dominant cations Ca>Mg>Na+K and dominant anions HCO₃>SO₄>Cl. These show an environmental and shallow origin according to the Cl-SO₄-HCO₃ ternary diagram (Kutlu, 2015). Accordingly, the geothermal waters have high sulphate contents, which might be attributed to gypsum and pyrite mineral phases in impermeable cap rocks. The waters are

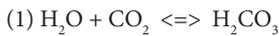
Table 1: In-situ measurements and hydrogeochemical analyses of the geothermal waters in Pamukkale and environs (Kıymaz, 2011; Kutlu, 2015; Uzun, 2017).

Sample	Location	T(°C)	pH	Eh (mV)	EC (µS/cm)	Na ⁺ (mg/l)	K ⁺ (mg/l)	Ca ⁺² (mg/l)	Mg ⁺² (mg/l)	B ⁺³ (mg/l)	F ⁻ (mg/l)	SO ₄ ⁻² (mg/l)	Cl ⁻ (mg/l)	Si ⁺⁴ (mg/l)	NO ₃ ⁻ (mg/l)	HCO ₃ ⁻ (mg/l)
EU-1	Plütonyum spring	34.8	6.61	155	2420	42.10	5.52	442	94.10	0.80	1.82	706	14.10	30.10	1.20	1176.1
EU-2	Gelin Hamamı	34.7	6.69	157	2410	42.40	5.60	434	90.40	0.90	1.40	661	14.60	30.50	1.10	1125
EU-3	Beltes spring	34.1	6.91	144.3	2410	42.50	5.45	445	96.10	0.80	1.35	662	14.70	30.40	1.00	1147.3
EU-4	Jan-darma spring	34.1	6.96	128.7	2410	42.5	5.45	325	95.50	0.80	1.34	661	12.90	30.50	0.90	1164.2
EU-5	Karahayıt spring	44	6.67	136.8	2540	117	24.3	367	118	1.60	1.85	915	38.8	29.70	5.60	1196.3
DK-1	Pamukkale spring	35	6.56	259.9	2430	44.2	5.45	99.4	401	1.0	0.8	649	13	15.8	1.56	1098
DK-2	Plütonyum spring	35.1	6.44	282.1	2400	40.1	5.08	95.4	479	0.9	1.8	642	14.1	15.2	1.88	1079.7
DK-3	Karahayıt spring	46.6	6.52	63	2680	107	21.1	124	414	1.6	2.2	905	31.8	26.8	0.65	927.2
AB-1	Özel İdare spring	35	6.22	210	2410	48.85	15.55	455.05	69.90	0.71	1.35	624.8	12.29	19.19	0.51	1128.5
AB-2	Jan-darma spring	33	6.24	229	2420	42.95	3.10	449.90	71.25	0.46	1.39	611.9	12.64	19.12	0.58	1159
AB-3	Karahayıt spring	52	6.39	113	2790	131.65	21.80	528.5	123.15	0.96	1.88	872.3	27.23	28.94	0.01	1189.5
AB-4	Karahayıt Richmond Hotel	48	6.18	161	2810	124.3	17.25	440.75	95.30	1.60	2.21	879.7	51.61	21.32	0.05	1128.5
AB-5	Karahayıt groundwater	23.9	8.01	287	448	86.4	0.65	3.77	0.48	0.22	0.34	11.12	5.57	5.10	13.53	231.8

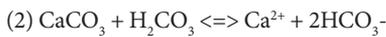
AB-6	Gölemezli well 1	67	6.89	194	2420	247.7	52.2	148.75	72	3.40	0.99	377.7	31.41	29.92	<0.01	1159
AB-7	Gölemezli well 2	69	6.69	144	3470	207.85	42.95	555.5	84.25	2.99	1.20	431.9	27.44	24.31	<0.01	2074
AB-8	Gölemezli Hamam	59	6.28	253	4460	431.6	45.05	464.15	109.5	5.74	2.45	1664	70.84	59.03	<0.01	1250.5

immature waters according to the ternary diagram of Na/1000-K/100-√Mg (Figure 6; Giggenbach, 1988). Moreover, geochemical thermometers of Na-K and Na-K-Ca show calculated temperatures ranging from 200 to 280 °C in the study area.

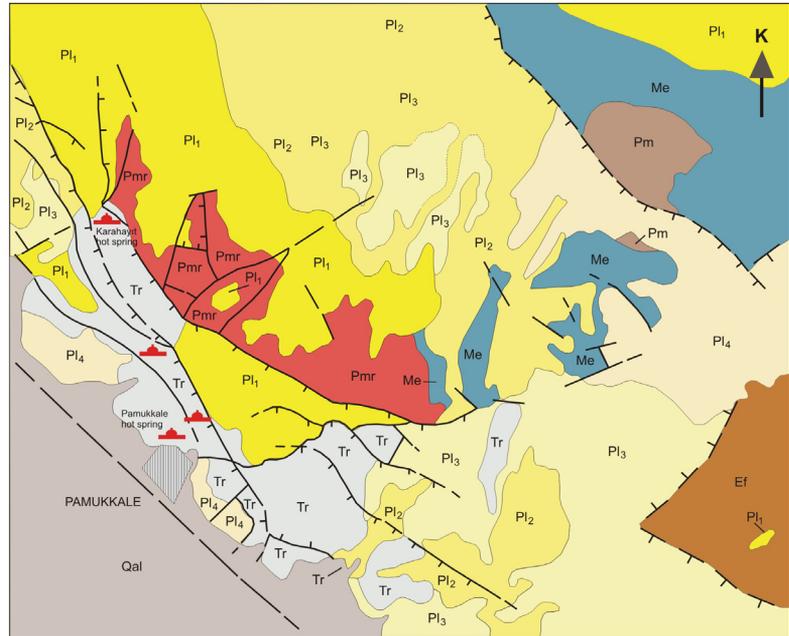
The abundant travertine deposits of Pamukkale are related to the unusual geological, tectonic and geomorphological setting in the study area (El Desouky *et al.*, 2015; Figure 7). The study area is characterised by abundant carbonate successions in its substratum, which provide the necessary parent carbonate sources for the formation of travertine deposits. According to El Desouky *et al.* (2015), Miocene to Pliocene sub-volcanic activities in the area probably play a major role in the formation of travertine deposits by (1) acting as a heat source for the geothermal fluids, (2) enhancing decarbonation processes in the deep subsurface and (3) contributing to the CO₂ source via mantle degassing. The extensional tectonic features associated with the development of the study area cause a network of faults and fissures that enhance circulation of geothermal waters. In the area, the rain fall rates of up to 600 mm and the presence of high mountains (1500–2000 m) ensure the necessary meteoric waters and hydraulic head for the travertine-precipitating geothermal waters. The formation of travertine deposits depends upon the solubility of CaCO₃ controlled principally by CO₂ partial pressure, temperature and pH values in which reaction equilibriums play an important role:



CO₂ is dissolved in waters as H₂CO₃



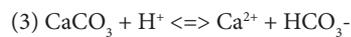
The process that increases the CO₂ proportion enhances the solution of CaCO₃, whereas each reduction in CO₂ proportion preludes the precipitation of CaCO₃. Under lower pH values, in which the most carbonates are solved as H₂CO₃, the reaction proceeds to the right side; under higher pH values, the reaction proceeds to the left side due to the precipitation of CaCO₃. CO₂ is less soluble in hot waters than in cold waters. The solubility of CaCO₃ decreases slightly with increasing temperatures.



EXPLANATION

CENOZOIC	Quaternary	Qal	Alluvium	Contact Fault Possible fault Hot spring
	Late Pliocene	Pl ₄	Tosunlar formation	
		Pl ₃	Kolonkaya formation	
	Early Pliocene	Pl ₂	Sazak formation	
Pl ₁		Kizilburun formation		
Eocene	Ef	Conglomerate, sandstone, marl (flysch)		
	Me	Limestone		
MESOZOIC		Pmr	Marble, quartzite, schist	
PALEOZOIC		Pm	Various schist	

Figure 4: Geological map of the geothermal field of Pamukkale and environs (modified from Akkuş *et al.*, 2005).



HCO₃⁻ ions are derived from the reaction of H⁺ with the carbonates.

For the formation of travertine deposits in Pamukkale and environs, the temperature and CO₂ partial pressure are two rival parameters. In Pamukkale, the important

parameter is the decrease of CO₂ partial pressure, and probably the temperature plays a secondary role. The strong temperature decrease in the ascending geothermal waters increases CaCO₃ solubility in waters. Moreover, the pressure release due to escape of CO₂ at the surface encourage carbonate precipitation. In volcanic activities in

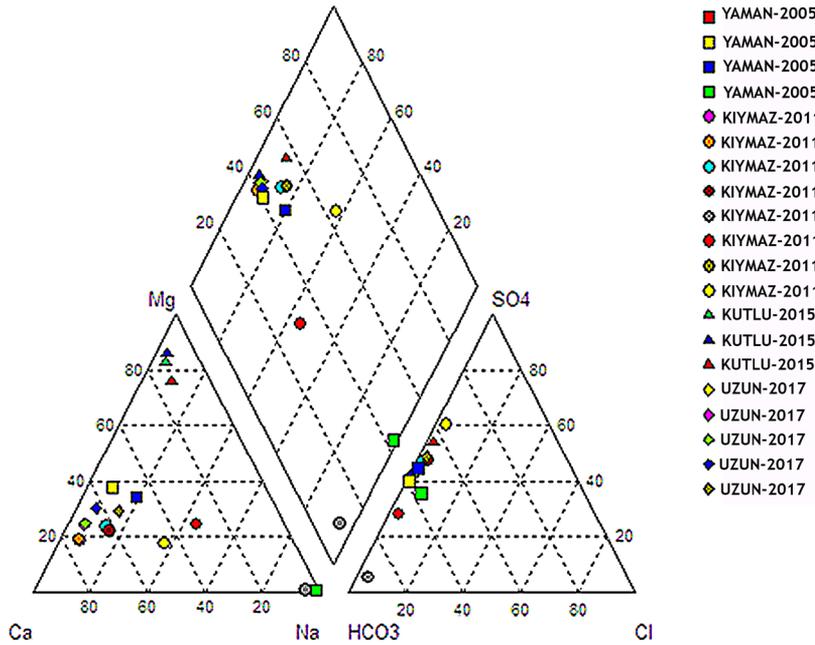


Figure 5: Piper diagram of the geothermal waters in Pamukkale and environs.

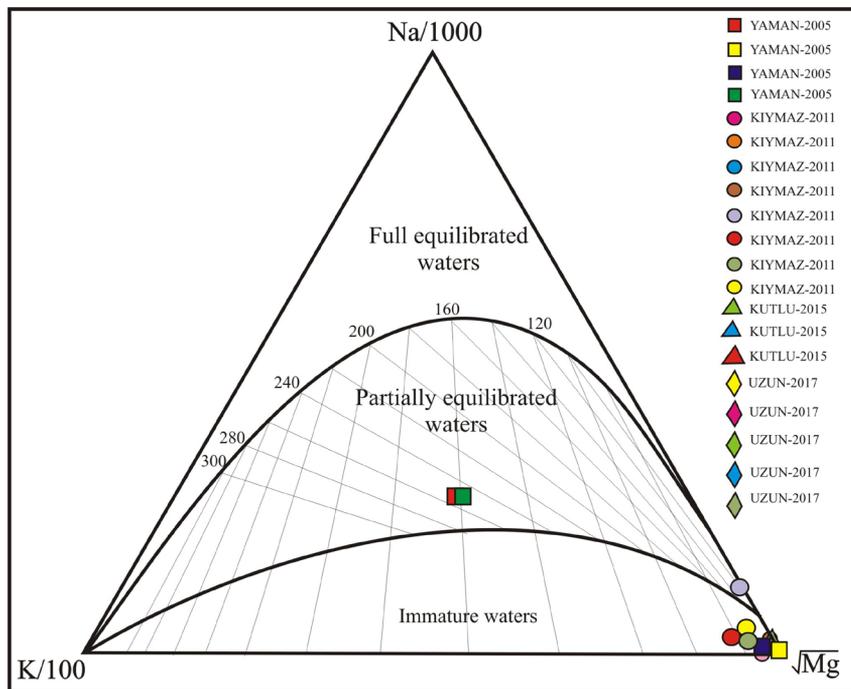


Figure 6: Na/1000-K-100-√Mg ternary diagram of the geothermal waters of Pamukkale and environs.

depth, i.e. volcanic rocks in Denizli (Semiz, 2003), the partial pressure of CO₂ in which CaCO₃ is solved is high. With the additional carbonate dissolution in the reservoir, CO₂ is consumed. However, the CO₂ partial pressure decreases insignificantly. Moreover, the geothermal waters are supersaturated due to CaCO₃ if the waters reach the reservoir. The carbonates precipitate if temperature equilibrium by the fast ascending geothermal waters does not take place in the same proportion as the pressure decrease at outflow. In addition, it is well known that

blue-green seaweeds are involved in carbonate precipitation: the seaweeds extract CO₂ from the system in the microenvironment and thus encourage carbonate precipitation as aragonite (Ramon, 1983).

In Pamukkale, the formation of travertine deposits was generated in five phases (Eşder and Yilmazer, 1991):

1. formation of the Çökelez fault, which strikes in NW-SE direction. The outflow of the geothermal waters is controlled by the faults directly. Today,

the travertine deposits of this phase are noticeable in high elevation spheres. For these travertine deposits of the first phase, a U-series age of 400 ka can be accepted (Altunel, 1996).

2. The formation of travertine deposits is of modern origin. An option of an outflow of hot spring was developed by the formation of the Karahayıt fault. The travertine of the second phase is widespread in accordance with foothill slope.
3. There is a stairway fault in the area. The travertine of this phase is observed in SE part of Pamukkale and was utilised in the construction of the ancient city Hierapolis.
4. In this phase, the stairway faults were generated widespread.
5. The last phase shows the landscape as it is nowadays. Great parts of the formation of travertine deposits in higher elevation areas have been eroded. Recent travertine forms modern carbonate precipitations as travertine consisting particularly of aragonite.

Isotope geochemistry

In the study area, the stable isotope compositions ($\delta^{18}\text{O}$ and $\delta^2\text{H}$) in the geothermal waters are shown in Figure 8 (Kıymaz, 2011; Kutlu, 2015; Uzun, 2017; Yaman, 2005). $\delta^2\text{H}$ values in geothermal waters vary between -61.9 and -51.8 ‰, whereas $\delta^{18}\text{O}$ values range from -9.23 to -5.84 ‰. The tritium contents of the geothermal waters vary between 0.7 and 3.3 TU. The samples of the geothermal waters of Pamukkale lie along the global meteoric water line (GMWL), whereas the samples of the high temperature geothermal waters in Kızıldere (Özgür, 1998; Yaman, 2005) deviate from GMWL, indicating intense water-fluid interaction under high temperature conditions. ^3H values up to 3.3 TU show an atmospheric or anthropogenic origin. Therefore, there is a mixing process between fresh groundwaters and geothermal waters in the area of Pamukkale.

Discussion

The geothermal waters of Pamukkale and environs were modelled hydrogeologically (Figure 9). The meteoric waters in the drainage area percolate at fault and fracture zones and through permeable clastic sediments

into the reaction zone of the roof area of a magma chamber. The chamber is located at a probable depth of 4-5 km, where meteoric fluids are heated by the cooling magmatic melt and ascend to the surface due to their lower density caused by convection cells. The volatile components of CO_2 , SO_2 , HCl , H_2S , HF , and He from the magma reach the geothermal water reservoir, where an equilibrium forms between altered rocks, gas components, and fluids. Thus, the geothermal waters ascend along the tectonic zones of weakness at the continental rift zones of the Menderes Massif in the form of hot springs, gases, and steams. These fluids are characterised by high to medium CO_2 , H_2S and NaCl contents. It is very important to note that the fluids indicate a reduced pH-neutral environment after equilibrium adjustment with hard rocks in the reaction zone, namely in the roof area of magma chamber (Giggenbach, 1992).

The formation of travertine deposits depends upon the solubility of CaCO_3 , controlled principally by CO_2 partial pressure, temperature and pH values, in which reaction equilibriums play an important role. Recent travertine deposits form the modern carbonate precipitations consisting of aragonite. In the study area, the travertine can be considered as terrace, ridge and channel type travertine (Altunel, 1996). Additionally, the geothermal waters of Pamukkale have high sulphate contents of up to 650 mg/l (Özgür *et al.*, 2004) and Rn concentrations of up to 20 Bq/l (Kulalı, 2016). These features are connected with the decay of uranium minerals in the metamorphic rocks of the Menderes Massif as in which the high sulphate contents are associated with sulphide contents such as pyrite as well as gypsum minerals in reservoir and cap rocks.

Acknowledgements

This study has been funded by the Scientific Research Coordination Office of the Suleyman Demirel University under contract numbers 4137-YL1-14 and 4494-YL1-15. We thank Mrs. Eda Aydemir, Mr. Ümit Memiş and Mr. Mehmet Arıcı (Süleyman Demirel Üniversitesi, Isparta, Turkey) for completion of figures of this paper. We would also like to thank Dr. Ali Gökğöz (Pamukkale University, Denizli, Turkey) for kindly examining and correcting the manuscript.



Figure 7: Sinter terrace of the travertine deposits in the study area of Pamukkale.

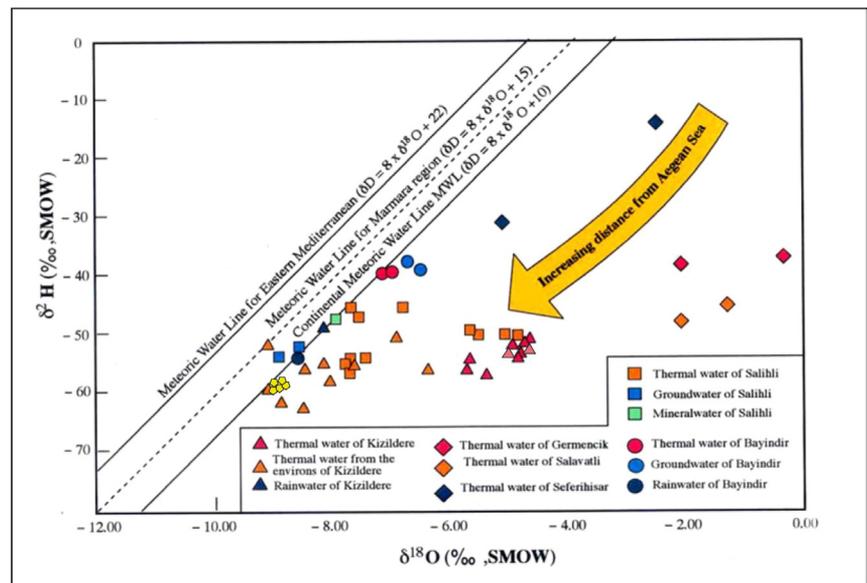


Figure 8: Plot of $\delta^{18}\text{O}$ versus $\delta^2\text{H}$ of the geothermal waters in Pamukkale and environs. For the data of stable isotopes see Özgür (1998), Yaman (2005) and Kıymaz (2011).

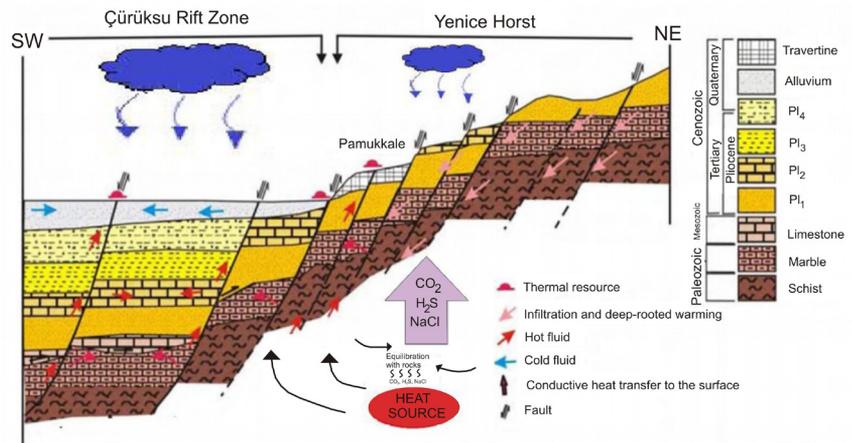


Figure 9. Hydrogeological modeling of the geothermal waters of Pamukkale and environs (P4: Tosunlar formation; P3: Kolonkaya formation; P2: Sazak formation; P1: Kızılburun formation) (modified from Dilsiz *et al.*, 2004).

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