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Geochemical and isotope characterization of geothermal spring waters in Sri Lanka: Evidence for steeper than expected geothermal gradients

Rohana Chandrajith^{a,*}, Johannes A.C. Barth^b, N.D. Subasinghe^c, Dirk Merten^d, C.B. Dissanayake^c

^a Department of Geology, Faculty of Science, University of Peradeniya, Sri Lanka

^b Lehrstuhl für Angewandte Geologie, GeoZentrum Nordbayern, Schlossgarten 5, D-91054 Erlangen, Germany

^c Institute of Fundamental Studies, Hantana Road, Kandy, Sri Lanka

^d Institut für Geowissenschaften, Friedrich-Schiller Universität, Burgweg 11, D-07749 Jena, Germany

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SUMMARY

Seven geothermal springs from the Precambrian high-grade metamorphic terrain of Sri Lanka were investigated to assess their formation processes and to determine reservoir temperatures based on their chemical compositions, Silica-based geothermometric calculations for the Marangala and Nelumwewa springs showed the highest average reservoir temperatures of 122 °C and 121 °C, respectively. Samples of low temperature (<35 °C) groundwater from nearby springs, piezometers and open wells were also collected for comparison. All samples were analyzed for their major and trace element compositions as well as stable isotope ratios ${}^{2}H/{}^{1}H$ and ${}^{18}O/{}^{16}O$ (expressed as $\delta^{2}H_{H2O}$ and $\delta^{18}O_{H2O}$). Discharge temperatures of the thermal waters varied from 39-62 °C. These waters showed low concentrations of selected trace elements (Fe < 0.09; Mn < 0.04; Cu < 0.01; Cr < 0.01; As < 0.025 mg/L)) and were also comparable to that of non-geothermal groundwaters. Stable isotope compositions of geothermal waters ranged from -6.5to -5.0% for $\delta^{18}O_{H2O}$ and between -39% to -28% for $\delta^{2}H_{H2O}$. In the non-geothermal waters, the isotope values were almost identical within the analytical uncertainties of 0.1‰ and 1‰ for $\delta^2 H_{H2O}$ and $\delta^{18}O_{H2O}$, respectively. In addition, all isotope ratios of geothermal and non-geothermal water samples scattered around the local meteoric water lines for the dry and intermediate climatic regions of Sri Lanka, thus indicating origin from precipitation without further influences of evaporation or water rock interaction. This similarity to the local meteoric water lines also makes influences of seawater an unlikely factor. Close matches of geochemical and isotope data from geothermal and corresponding non-geothermal waters confirm the hypothesis of a common source. The proposed model for Sri Lanka subsurface waters is that rainfall from the dry and/or intermediate climatic zones percolates with little time delay downward through structurally weaker zones in high-grade metamorphic rocks. They are subsequently heated by steep or heterogeneous geothermal gradients that are most likely associated with the Highland-Vijayan thrust zone.

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1. Introduction

Although geothermal systems are commonly associated with volcanic activities or recent uplifts (Diamond and Harris, 2000), several hot water springs with low to intermediate temperatures can be found in the Proterozoic crust of Sri Lanka where recent volcanic or tectonic activities are unknown. Most of these wells discharge hot water as natural springs. In addition, an artesian hot water well occurs in one location. Measured surface temperatures of the geothermal springs in Sri Lanka ranged between 34 and 55 °C (Dissanayake and Jayasena, 1988). Recently some of these springs have fallen dry or were inundated in man-made reservoirs

and nowadays only seven geothermal springs occur in the highgrade metamorphic terrain of Sri Lanka. Only few such geothermal springs were reported in high grade terrains such as in India and Canada (Chandrasekharam and Antu, 1995; Jessop et al., 1991).

In recent years, these geothermal springs raised much interest for potential development of geothermal exploitation for green energy. Despite this interest, hardly any work has been published on the geothermal springs in Sri Lanka, except for the work of Dissanayake and Jayasena (1988) and Senaratne and Chandima (2011). These workers found that subsurface temperatures of some of the thermal springs are around 143 °C. These temperatures raise speculation about economic use and with this about the origin of the waters. The localization of heat sources need further investigation as they are located in a Precambrian high grade metamorphic terrain that is untypical for thermal springs. Recently, Senaratne and Chandima (2011) discussed the origin of the Marangala



^{*} Corresponding author. Tel.: +94 81 2394218, mobile: +94 77 7812392; fax: +94 81 2388018.

E-mail addresses: rohanac@hotmail.com, rohanac@pdn.ac.lk (R. Chandrajith).

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geothermal spring (Fig. 1) with structural and surface geological evidences. These authors outlined the mechanisms of the artesian type of well in the area. Although the hot springs of Sri Lanka are known since centuries, the origin and the structure of these hydrothermal systems are still not well understood. The main objective of this work was therefore to characterize geothermal springs in Sri Lanka by chemical and water isotope methods. Comparison with nearby low-temperature groundwaters and with precipitation helped to trace hydrogeological flow paths and to understand the recharge sources as well as to narrow down potential heating mechanisms.

With this, the present study interprets the first set of geochemical data and water stable isotope values (expressed as $\delta^2 H_{H20}$, $\delta^{18}O_{H20}$) that were obtained from the geothermal spring systems from the high-grade metamorphic terrain. These parameters have become important tools in hydrogeology and have widely been used

as natural tracers (Clark and Fritz, 1997). For instance, they were applied in geothermal systems with different origins (Delalande et al., 2011; Han et al., 2010; Pasvanoğlu and Chandrasekharam, 2011).

2. Geological and hydrological setting

The Precambrian basement of Sri Lanka is a small fragment of the Gondwana supercontinent, over 90% of which is made up of amphibolite to granulite grade metamorphic rocks. The remainder is covered by sedimentary sequences that belong to Jurassic, Miocene and Holocene periods (Fig. 1). The Proterozoic crust of Sri Lanka is divided into three major lithological units based on geochronologic, petrologic and geochemical data. These are known as the Highland Complex (HC), the Vijayan Complex (VC) and the Wanni Complex (WC)(Cooray, 1994). The folded belt of the HC is



Fig. 1. Map of the general geology (simplified from Cooray (1994)), topography, climatic zones and distribution of hot water springs in Sri Lanka.

met by the VC in the east and the WC in the west. The Highland Complex consists mostly of granulites and is dominated by rocks of granitoid composition with ages ranging from 1942 Ma to 650 Ma (Kröner et al., 1991). The amphibolite grade VC and WC show ages between 1100 Ma and 550 Ma (Kröner et al., 1991). The boundary between the HC and the VC is well defined as shown by the differences between metamorphic grades, structure and isotopic ages (Kröner et al., 1991). It has at least two tectonic blocks with marked lithological and geochemical differences. This boundary is one of the most interesting and controversial geologic features of Sri Lanka. It is considered to be a mineralized belt (Dissanayake, 1985) or a thrust zone (Kröner et al., 1991).

The hot water springs in Sri Lanka are located around or close to the boundary between HC and VC. Lithologically, meta-granites, granitic gneisses and quartzites underlie the region while some scattered bodies of dolerite dikes can also be observed. Recent volcanism is an unlikely source of heat as the youngest igneous activity in Sri Lanka had occurred in Jurassic or late Cretaceous times (Takigami et al., 1999; Yoshida et al., 1989). All geothermal springs are located in the hard rock terrain where limited residual primary porosity occurs but ductile and brittle fractures, joints and fault planes as well as bedding surfaces provide a network of interconnected conduits for groundwater movement. Some of these features extend as lineaments for several kilometers and produce secondary porosity and permeability conditions for storage and circulation of deeper ground waters.

The Island of Sri Lanka is characterized by a humid and tropical climate in which monsoon rainfalls from the northeast and southwest control the majority of hydrologic and agro-ecologic differences. Regional differences in precipitation are large and the island is spatially divided into a dry zone, an intermediate zone and a wet zone (Fig. 1). The wet zone comprises the southwestern part of Sri Lanka, where 50% of the year has rainfall intensities from 2280 to 5100 mm per year. The dry zone occupies two-thirds of the island and receives about 1000 mm of annual rains that originate entirely from the northeast monsoon. The intermediate climatic zone receives mean annual rainfall of 1700 mm. The humid climate in Sri Lanka is characterized by seasons, in which potential evaporation exceeds precipitation (Domrös, 1979). Interestingly, all hot water springs are located in the dry and intermediate zones that are characterized by a semi-arid climate. The high ambient temperatures and prevailing winds significantly contribute to significant evaporation and transpiration losses in the region.

All thermal water springs have variable flow rates and discharge temperatures (Table 1). The geothermal springs are also located in the lower altitude regions (<100 m) of the island, except for the

spring at Marangala (Fig. 1) that has an altitude of about 100 m above sea level. In addition, the Marangala thermal well, which was drilled in 1983 to a depth of 21 m below ground level in a SW-NE running lineament, constitutes an artesian well with a flow rate of 370 L/min and a pressure head of about 3 m above the ground.

3. Materials and methods

Anions, cations and isotope ratios of water (expressed as $\delta^{18}O_{H2O}$ and $\delta^{2}H_{H2O}$) from seven hot water springs, three coldwater springs, and four deep wells were collected for this study (Table 1). Out of these, one cold-water spring is located close to the hot water springs and all other cold groundwater-sampling sites were selected from the nearest possible location (<100 m) to the sampled thermal springs. The Nelumwewa geothermal spring (NW in Fig. 1) is located in a lake and the cold-water sample was taken from a deep borehole situated approximately one kilometer away from the hot spring. Water temperature, pH, electrical conductivity (EC) and alkalinity were measured *in situ*. Several aliquots of samples were collected from each location with field blanks and duplicates.

Water samples collected for geochemical analyses were filtered through 0.45-µm syringe filters and aliquots of the filtered water were collected into HDPE vials as unacidified samples for anion analyses and in a separate vial acidified with ultrapure nitric acid to pH < 2 for major and trace cation analyses. Unfiltered, 25 mL of sample were collected into screw capped HDPE tubes for oxygen and hydrogen isotope ratio analyses of the water. The alkalinity of samples was determined by titration with 0.16 N H₂SO₄ to a pH of 4.3 using a Hach[®] digital titrator. Hach SensION[®] portable meters and electrodes were used for in situ measurements of pH, EC and temperature. Dissolved silica contents in samples were measured using the silico-molybdate method (Eaton et al., 2005). This method reacts water samples with acid ammonium molybdate reagents and the resultant color was subsequently measured with a Hach® DR 2400 visible spectrophotometer at 815 nm. Major anions (Cl-, NO_3^- , SO_4^{2-} and F^-) were determined using ion chromatograph (Dionex ICS 2000) with a Dionex AS11HC column. Major cations and trace metals in water samples were determined by inductively coupled plasma-optical emission spectrophotometry (ICP-OES; Varian 725ES).

Measurements of oxygen and hydrogen isotope ratios in water samples were carried out by wavelength-scanned cavity ringdown infrared spectroscopy with a Picarro L1102-*i* instrument that

Table 1

Background information on sampling sites (* temperatures of non-geothermal water depend on the ambient temperature; water temperature >35 °C is considered as hot groundwater).

Site	Code	Geographic coo	rdinates	Altitude (masl)	Temp.* (°C)	Description
		N	E			
Mahapelessa	TW1	6°14.49′	80°59.04′	32	45.5	Hot water spring
Mahapelessa	GW1	6°15.14′	80°59.02′	32	29.0	Cold water spring
Rankihiriya	TW2	8°42.27′	80°59.31'	65	39.1	Hot water spring
Rankihiriya	GW2	8°41.14′	80°59.35′	65	31.0	Cold water spring
Kanniyai	TW3	8°36.16′	80°19.17′	34	41.7	Hot water spring
Kanniyai	GW3	8°36.13′	80°18.22′	34	31.2	Deep dug well
Nelumwewa	TW4	7°53.22′	81°12.06′	45	62.2	Hot water spring
Nelumwewa	GW4	7°54.80′	81°11.76′	45	30.4	Borehole well
Mahaoya	TW5	7°33.08′	81°21.11′	45	53.5	Hot water spring
Mahaoya	GW5	7°33.07′	81°21.21′	45	33.1	Borehole well
Marangala	TW6	7°21.42′	81°19.33′	102	43.4	Hot water spring
Marangala	GW6	7°22.32′	81°20.17′	103	31.5	Borehole well
Kapurella	TW7	7°38.02′	81°25.08′	30	58.0	Hot water spring
Bibile	GW7	7°10.30′	81°13.50′	305	29.4	Cold water spring

was coupled with a vaporization module. Results are reported in the delta (δ) notation versus the V-SMOW standard and expressed in per-mille (∞). External reproducibility was determined with control standards and was better than 0.1 and 1 ∞ (1 σ) for δ^{18} O and δ^{2} H, respectively. The ambient meteoric isotope data in Sri Lanka were obtained from the Isotope Hydrology Information System (ISOHIS) database of the International Atomic Energy Agency/ World Meteorological Organization (IAEA/WMO, 2006). The ISOHIS database has recorded δ^{18} O_{H2O} and δ^{2} H_{H2O} values for the precipitation collected from 12 stations in Sri Lanka from 1986 to 1995 (http://isohis.iaea.org; accessed on 15 September 2011).

Mineral saturation indices were calculated for both geothermal and non-geothermal water using PHREEQC version 2 (Parkhurst and Appelo, 1999) that is designed to perform a wide variety of low-temperature aqueous geochemical calculations.

4. Results and discussion

4.1. Geochemistry of thermal waters

Descriptions of sampling locations and geochemical results of thermal and non-geothermal waters are listed in Tables 1 and 2. The discharge temperature of geothermal waters varied from 39.1 °C (Rankihiriya) to 62.2 °C (Nelumwewa) while their pH varied from 5.7 (Kanniyai) to 8.0 (Kapurella). The electric conductivity (EC) of geothermal waters varied from 532 μ S/cm (Kanniyai) to 7360 μ S/cm (Mahapelessa) while it ranged from 354 μ S/cm (Bibile) to 7290 μ S/cm (Mahapelessa) in non-geothermal groundwaters (Table 2). This indicates a low mineralization of geothermal and non-geothermal waters except at the Mahapelessa springs.

The major ion distributions in both types of waters are shown as Stiff plots in Fig. 2. Sodium is the dominant cation while chloride and sulfates are the main anionic constituents in both hot and cold water samples. Geothermal and non-geothermal waters from the same region show remarkable similarities in their pH, EC and in major ion contents. This indicates common recharge sources and circulation patterns, except for the samples from Nelumwewa spring, where the deep borehole sample was taken approximately one kilometer away from the hot water spring. Nevertheless, the cold-water spring from Bibile, shows higher bicarbonate contents that is compensated by lower concentrations of all other major elements. The Na⁺, Cl⁻ and HCO₃⁻ concentrations in geothermal waters range from 15.8 to 1128 mg/L, 13.8 to 2384 mg/L and 7.8 to 184 mg/L, respectively. The total dissolved silica content of geothermal water ranged from 20.8 mg/L to 42.2 mg/L and from 20.3 mg/L to 44.2 mg/L for non-geothermal waters. A higher concentration of dissolved silica is usually found in high-temperature geothermal waters, particularly in those associated with volcanic systems.

Based on the major element composition, hot and cold groundwater can be classified into three groups as Na–Cl–HCO₃-type (Mahapelessa), Na–Cl–SO₄-type (Nelumwewa, Mahaoya, Marangala and Kapurella) or Ca–Cl–SO₄-type (Rankihiriya and Kanniyai). These are in contrast to sodium–HCO₃ waters that have been described as common in geothermal systems associated with metamorphic terrains as indicated by Vengosh et al. (2002). Geothermal springs located in the northeastern part (Rankihiriya and Kanniyai) of Sri Lanka are mostly dominated by high bicarbonate contents and Özler (2000) indicated that the HCO₃⁻ content can increase with time and travel distance underground. However, both these springs showed lower discharge temperatures compared to other geothermal springs in Sri Lanka.

The springs in the middle part of Sri Lanka (i.e. Nelumwewa, Mahaoya, Marangala and Kapurella) are characterized by highsulfate water, while the Mahapelessa spring in the south of the country is characterized by higher chloride–sodium contents. The Mahapelessa spring which is located away from other geothermal springs is well-mixed with mineralized waters. This is indicated by

Table 2

Concentrations of major and trace chemica	l constituents (mg/L) in geothermal	and non-geothermal water	(HD – total hardness: nd – not detected)
			· · · · · · · · · · · · · · · · · · ·

				-		-								
	Location	pН	EC (µS/cm)) HCO	₃ HD	SO_4	Cl	PO ₄	NO_3	F	SiO ₂	Na	K	Ca
Hot water springs	Mahapelessa	7.11	7360	7.80	nd	131	2384	0.10	3.2	2.62	68.8	1128	39.5	669
	Rankihiriya	6.60	998	184	246	2.61	13.80	0.50	1.4	0.23	90.7	15.80	5.98	76.2
	Kanniyai	5.72	532	93.8	106	2.16	13.78	0.29	1.1	0.12	52.7	17.10	5.69	54.0
	Nelumwewa	8.00	1288	35.2	45.) 231	222.3	0.16	1.2	5.38	96.2	273.6	10.6	27.7
	Mahaoya	7.14	1261	51.2	114	417	74.33	0.17	0.6	5.11	72.3	249.6	11.3	67.9
	Marangala	7.30	1210	50.7	96.) 465	26.84	0.16	0.7	5.10	98.7	245.3	11.8	62.2
	Kapurella	8.40	1403	82.9	nd	184	301.9	<0.20	<0.1	5.95	81.9	280.3	8.71	20.3
Cold groundwater	Mahapelessa	7.20	7290	8.30	nd	130	2141	0.05	3.2	2.79	68.9	1132	39.1	713
	Rankihiriya	6.16	653	125	152	4.14	24.74	0.38	0.9	0.24	84.4	17.40	4.64	52.8
	Kanniyai	6.09	453	119	150	1.29	22.42	0.10	1.1	0.15	44.6	19.60	4.27	81.5
	Nelumwewa	6.25	963	125	358	18.6	178.8	0.40	2.6	0.34	90.3	37.40	2.20	182
	Mahaoya	7.14	1229	56.0	118	416	74.20	0.36	0.6	5.34	39.4	253.2	12.6	74.4
	Marangala	6.73	593	92.7	71.) 126	14.49	0.52	1.0	2.50	93.3	105.2	6.22	40.3
	Bibile	7.02	354	250	190	3.20	5.380	0.17	0.0	0.70	85.0	16.90	3.61	89.0
	Location	Mg	Ba	Sr	В	Li	Fe	Mn	Cr	Cu		Al	Zn	As
Hot water springs	Mahapelessa	2.69	0.75	16.0	0.39	0.34	0.086	0.027	0.006	0.00	8	0.13	0.56	<0.025
	Rankihiriya	38.4	2.29	0.12	0.40	0.01	0.058	0.001	0.004	0.00	8	0.13	0.62	< 0.025
	Kanniyai	7.54	1.05	0.13	0.32	0.01	0.032	0.001	0.002	0.00	9	0.07	0.63	< 0.025
	Nelumwewa	0.19	0.81	0.44	0.36	0.18	0.078	0.003	0.004	0.00	5	0.15	0.56	< 0.025
	Mahaoya	2.43	0.03	0.84	0.11	0.16	< 0.002	0.036	< 0.002	<0.0	02	<0.01	0.15	< 0.025
	Marangala	7.47	1.06	0.13	0.33	0.01	0.052	0.001	0.003	0.00	7	0.07	0.58	< 0.025
	Kapurella	0.31	0.16	0.40	0.12	0.17	0.040	0.010	< 0.002	0.00	8	0.03	0.02	<0.025
Cold groundwater	Mahapelessa	5.87	0.77	14.6	0.43	0.33	0.085	0.052	0.005	0.01	2	0.14	0.60	<0.025
	Rankihiriya	22.7	1.42	0.09	0.37	0.01	0.100	0.014	0.005	0.00	8	0.13	0.61	<0.025
	Kanniyai	10.3	1.98	0.22	0.34	0.03	1.647	0.222	0.005	0.00	7	0.15	0.57	<0.025
	Nelumwewa	14.4	0.93	0.99	0.16	0.09	0.129	0.711	0.003	0.13	3	0.10	0.36	< 0.025
	Mahaoya	2.24	0.50	0.81	0.30	0.15	0.209	0.077	0.004	0.00	5	0.16	0.52	< 0.025
	Marangala	4.58	0.50	0.11	0.34	0.08	0.158	0.185	0.004	0.01	1	0.16	0.62	< 0.025
	Bibile	11.1	1.06	0.17	0.28	0.01	0.082	0.020	0.004	0.00	7	0.10	0.55	< 0.025



Fig. 2. Stiff plots of water samples from geothermal springs (left) and corresponding non-geothermal water (right), except Kapurella (KR) hot water spring and cold-water spring of Bibile (BL).

high chloride–sodium composition and also by high EC (7360 μ S/ cm) values. Both geothermal and non-geothermal waters from Mahapelessa are also characterized by higher concentration of alkali and alkaline earth elements when compared to those of other hot or cold springs.

At first sight, the chloride-sodium character might be related to mixing with sea water. However, the springs are located 40 km away from the ocean and sea water intrusion is unlikely because of impermeable metamorphic rocks. Isotope mass balances between sea water (δ^{18} O = -0.04 permille) cold and hot groundwaters with -5.07 and -4.97 permille respectively showed that sea water can only constitute a minor contribution if anything to these waters. Interestingly, the groundwater in the nearby terrains is also characterized by high amounts of dissolved solids, however with more saline characters. The Na/Cl and other alkali and alkaline earth metal ratios have been used to identify the mechanism for acquiring salinity and saline water intrusion in semi-arid regions (Diaw et al., 2012; Sami, 1992; Su et al., 2009). The Na/Cl and Mg/Ca ratios of the Mahapelessa hot spring are 0.47 and 0.005 while 0.53 and 0.008 for the cold water spring; hence the water is rather altered by mixing with high saline water in the nearby terrain during vertical migrations. Even shallow well water in the nearby terrain also showed higher EC values ranging from 450 to 1220 μ S/cm with an average of 690 μ S/cm (Shortt et al., 2003)

With a few exceptions, cold-water springs and deep groundwaters show almost identical trace element variation patterns with their corresponding geothermal waters (Table 2). In contrast, Fe- and Mn-contents are slightly higher in cold groundwaters compared to nearby hot springs. High levels of fluoride, up to 5.95 mg/L were observed in samples particularly in thermal springs located in the middle part of the country while nearby non-geothermal waters also showed similar enrichments of fluoride. The alkaline environment favors the desorption of fluoride from mineral phases (Saxena and Ahmed, 2001) such as micas and amphiboles that are dominant in high-grade metamorphic rocks. This is similar to findings from a previous groundwater geochemical survey carried out in Sri Lanka that showed anomalous enrichment of fluoride and some base metals such Cu, Zn, V and Co along the HC-VC boundary of Sri Lanka (Dissanayake and Weerasooriya, 1986). The silica content of thermal springs ranges from 53 to 99 mg/L while cold springs and wells have silica content ranging from 40 to 93 mg/L. The arsenic content in all studied samples was lower than 0.025 mg/L.

4.2. Hydrogeochemical thermometry

In the last few decades, various geothermometers were introduced to calculate reservoir temperatures of geothermal waters based on the chemical constituents (Arnorsson, 1983; Fournier, 1977, 1979; Giggenbach, 1988; Kharaka and Mariner, 1989; Truesdell and Fournier, 1977; Verma and Santoyo, 1997). Among these, the silica (quartz, chalcedony, amorphous silica etc.) and alkali geothermometers (Na–K, Na–Li, Na–K–Ca etc.) are the most commonly applied. These temperature equations, based on empirical or semi-empirical laws, rely on specific equilibrium constants that refer to specific mineral-solution reactions in geothermal reservoirs. Silica-quartz/chalcedony-based geothermometers are widely applied to calculate the temperature of low enthalpy reservoirs (Fournier, 1977, 1979; Giggenbach, 1988; Truesdell and Fournier, 1977). Most of these geothermometers were based on water–rock equilibrium in geothermal reservoirs in volcanic geothermal terrains. The Na/K ratio and Na–K–Ca geothermometers proposed by Giggenbach (1988) can also be used to calculate the possible equilibrium temperatures. Na–K geothermometer is mostly applicable to reservoir temperatures above 180 °C and may yield erroneous values for low temperature waters (Arnorsson, 1983) while Na–K–Ca geothermometer is more suitable for waters with high Ca contents (Fournier and Potter, 1979). Fouillac and Michard (1981) introduced statistical Na–Li thermometric relationships for geothermal waters that mainly depend on the fluid salinity and on the reservoir type. These relationships have been described to yield more reliable reservoir temperatures of geothermal water due to low reactivity of Li (Kharaka and Mariner, 1989).

The reservoir temperatures for the studied geothermal springs in Sri Lanka were evaluated by silica based (conductive cooling, maximum steam loss and chalcedony) and cation based (Na–Li) geothermometers (Table 3). These geothermometers yielded wide ranges of reservoir temperatures for the same hot spring, possibly due to errors in the coefficient of geothermometric equations (Du et al., 2005; Verma and Santoyo, 1997). The modified silica geothermometer of Verma and Santoyo, (1997) yielded the highest reservoir temperatures that range from 105 °C (Kanniyai) to 138 °C (Marangala) for all geothermal springs in Sri Lanka.

For comparison, the silica–quartz conductive cooling and steam loss temperature equation proposed by Fournier (1977) gave reservoir temperature ranges from 97 °C (Kanniyai) to 132 °C (Nelumwewa). Na–Li geothermometric calculations also indicated promising and consistent reservoir temperature that are comparable to silica based geothermometric calculations with temperatures between 96 °C (Mahapelessa) and 129 °C (Nelumwewa). The average reservoir temperature of geothermal springs that were calculated from all methods (Table 3) varies from 88 °C (Kanniyai) to 120 °C (Nelumwewa). These calculated reservoir temperatures also correlate well with the surface discharge temperatures of each spring.

Due to possible mixing in geothermal waters in Sri Lanka with surrounding groundwater, the Na–K–Mg geothermometer according to Giggenbach (1988) cannot be applied reliably. This mixing was also evident from the modified Na–K–Mg ternary diagram drawn for the geothermal waters of Sri Lanka (Fig. 3) (Giggenbach, 1988; Shevenell and Goff, 1995) that indicates disequilibrium with their associated host rocks. Most of the thermal spring waters plot away from the full equilibrium line and show immature or multiple mixing characteristics. However, partially disequilibrated water shows apparent reservoir temperatures of over 100 °C. Another geothermometer is based on dissolved silica and K–Mg contents (Giggenbach, 1988; Giggenbach and Glovert, 1992), in which a full equilibrium line is defined by plotting the logarithms of silica content versus K^2/Mg (Fig. 4) also shows lower reservoir temperatures of 45–125 °C for geothermal waters from the high-grade terrain. Based on the modified silica geothermometer (Verma and Santoyo, 1997), the hot spring from Marangala showed a maximum reservoir temperature of 138 °C, it only required 450 kJ/kg energy to heat the water, thus indicating their low enthalpy. As shown by all geothermometric calculations performed here, maximum reservoir temperatures are observed in the middle part of the geothermal zone. A prominent negative Bouguer gravity anomaly has also been recorded in the same area (Hatherton et al., 1975).

Results of thermodynamic mineral equilibrium calculations are given in Table 4. Most of the secondary mineral phases such as goethite, haematite and illite are apparently supersaturated while quartz and chalcedony are saturated whereas amorphous silica is slightly under-saturated. The results suggest similar rock-water interactions for both geothermal and non-geothermal waters. The thermodynamic calculations also showed that all geothermal waters were close to saturation with quartz (Table 4), possibly due to conductive cooling. Fournier (1983) showed that the quartz equilibrium can be achieved at temperatures as low as 70 °C whereas below that the equilibrium with chalcedony will control the dissolution of silica. As shown in Table 4, most of the thermal waters have quartz near or slightly above the saturation limit for equilibrium. This indicates that the original silica content is preserved and does not re-equilibrate during circulation. Such stability may be attributed by fast flowing water through fracture zones in the subsurface (Cinti et al., 2009) and heat loss while traveling through cool rock.

Nevertheless, some uncertainty remains as the mechanisms of soluble mineral-based geothermometers for thermal water in high-grade metamorphic terrains may operate differently compared to waters from tectonically active or volcanic terrains. However, the majority of geothermometric equations were derived for high temperature geothermal water, associated with volcanic activities while chemical modification of geothermal water is limited in metamorphic terrain where no deep hot source rocks are involved.

It is interesting to observe that geochemical compositions of natural shallow groundwaters in nearby terrains closely match those of the hot groundwater. This indicates that all waters investigated should stem from a common circulation system. The absence of marked differences between hot and cold groundwaters also questions the investigated empirical geothermometric equations because the required equilibria that are the basis for all

Table 3

Geothermometric calculations using silica content of geothermal waters from Sri Lanka (MP – Mahapelessa; RK – Rankihiriya; KY – Kanniyai; NW – Nelumwewa; MO – Mahaoya; MG – Marangala; KR – Kapurella).

Geothermometer	Calculated temperature (°C)										
Thermal spring	MP	RK	KY	NW	МО	MG	KR				
Silica-quartz conductive cooling ^a	116	131	97	132	131	97	126				
Silica quartz max. steam loss ^b	114	127	99	128	128	98	124				
Silica-Chalcedony ^c	76	92	57	92	92	57	87				
Silica-Chalcedony ^d	87	103	69	103	103	68	98				
Modified silica geothermometer ^e	118	133	105	137	121	138	128				
Na-Li ^f	96	99	103	129	127	122	124				
Arithmetic mean (°C)	101	114	88	120	117	97	115				
±SD (°C)	17	18	20	18	16	31	17				

^a Temperature estimated by quartz geothermometer (Fournier, 1977): $t = \frac{1390}{5.19 - \log S} - 273.15$, where S = SiO_{2(aq)} in mg/L.

^b Temperature estimated by quartz geothermometer after steam loss (Fournier, 1977): $t = \frac{1522}{5.75 - \log S} - 273.15$, where S = SiO_{2(aq)} in mg/L.

^c Temperature estimated by chalcedony geothermometer (Fournier, 1977): $t = \frac{1000}{4.78 - \log S} - 273.15$, where S = SiO_{2(aq)} in mg/L.

^d Temperature estimated by chalcedony geothermometer based on field study (Arnorsson, 1983): $t = \frac{1112}{10-1005} = 273.15$, where S = SiO_{2(aq)} in mg/L.

^e Temperature estimated by modified silica geothermometer (Verma and Santoyo, 1997): $t = -44.119 + 0.24469S - 1.7414 \times 10^{-4} + 79.305 \log S$, where S = SiO_{2(aq)} (<295 mg/L).



Fig. 3. Distribution of geothermal spring water in modified Na/400-K/10 and \sqrt{Mg} (in mg/L) ternary diagram (after Giggenbach (1988) and Shevenell and Goff (1995)).



Fig. 4. A plot of log SiO_2 against the logarithm of the K^2/Mg ratio for the seven geothermal water samples from Sri Lanka on amorphus silica and quartz geothermometer equations (Giggenbach and Glovert, 1992).

geothermometric considerations may not have been reached due to faster circulation in a relatively fast conduit fracture system. Therefore, while reservoir temperatures may have been higher at larger depths the minimum temperatures for reservoir can be assumed to be those that were measured at the surface.

4.3. Isotope characteristics

Stable isotope values of geothermal waters are powerful geochemical tools for determination of the origin, nature, distribution and interactions of fluids in geothermal systems. They usually can only be modified by evaporation and mixing in shallow aquifers and by isotopic exchange with minerals in geothermal systems (Kharaka and Mariner, 2005; Truesdell et al., 1977). The $\delta^2 H_{H20}$ and $\delta^{18}O_{H2O}$ relationships of the geothermal and non-geothermal waters from Sri Lanka are presented in Table 5 and illustrated in Fig. 5 together with the regional and global meteoric water lines and ocean water collected from the Indian Ocean. The $\delta^2 H_{H2O}$ values in geothermal waters vary from -39% (Kanniyai) to -28% (Mahapelessa) whereas the $\delta^{18}O_{H2O}$ varies from -6.5% (Kanniyai) to -5.0% (Mahapelessa). Similar to the geochemical compositions, the isotope data from thermal and nearby non-geothermal waters are almost identical and thus confirm the hypothesis of a common source of recharge and common internal circulation patterns. The geothermal water from Mahapelessa and Kapurella springs are slightly enriched (>-5.0% for $\delta^{18}O_{H2O}$ and >-35% for $\delta^{2}H_{H2O}$) in the heavier isotopes compared to other geothermal springs, while water from the Nelumwewa, Mahaoya and Marangala show moderate enrichment (~-5.5% for $\delta^{18}O_{H2O}$ and ~-35% for $\delta^{2}H_{H2O})$ of heavy isotopes compared to that of Rankihiriya and Kanniyai springs in the north end of the geothermal zone.

Average $\delta^{18}O_{\rm H2O}$ values for geothermal and non-geothermal springs are -5.6 and -5.7%, respectively and with the analytical error of 0.1 can be regarded as similar. In general, geothermal waters are expected to have more negative average $\delta^2 H_{\rm H2O}$ and $\delta^{18}O_{\rm H2O}$ ratios than shallower non-geothermal groundwater water or surface water (Clark and Fritz, 1997) due to evaporation processes. Therefore, the slight enrichment of $\delta^{18}O_{\rm H2O}$ in geothermal waters from Mahapelessa and Kapurella is possibly an effect of evaporation during the discharge in which both springs also yielded higher reservoir temperatures.

The isotope characteristics of the geothermal and non-geothermal waters are also compared with data for ambient meteoric water that was monitored by the International Atomic Energy Agency (IAEA) in Sri Lanka as a part of the Global Network for Isotopes in Precipitation (GNIP). Although ideal comparison would be with meteoric water collected from a site near the spring over the same time period as sampling, such data are not available;

Table 4						
Results of thermodynamic equilibrium calculations using P	HREEQC v. 2.18	(Parkhurst and Appelo,	, 1999) (sample	e codes are as	given in Table 1).

	Saturati	on Index												
	TW1	TW2	TW3	TW4	TW5	TW6	TW7	GW1	GW2	GW3	GW4	GW5	GW6	GW7
Albite	0.09	-0.90	-2.16	-0.88	-1.66	-2.35	079	2.02	0.53	-0.19	0.83	0.64	1.51	0.53
Anorthite	-0.56	-0.76	-2.03	-1.77	-3.59	-2.40	-2.70	0.52	0.05	-0.18	0.31	-0.52	0.17	0.14
Barite	0.52	-0.04	-0.34	1.12	-0.11	1.60	0.34	0.72	0.13	-0.24	0.42	1.32	1.07	-0.12
Ca-montmorillonite	3.55	2.99	0.91	0.88	-1.31	0.68	-2.35	6.58	5.17	4.56	5.21	4.22	5.64	5.01
Calcite	-1.15	0.45	0.14	-0.86	-0.21	-0.41	0.87	-1.09	0.20	0.25	0.59	-0.34	-0.17	0.08
Chalcedony	0.05	0.23	-0.09	0.01	0.09	-0.11	0.23	0.58	0.63	0.35	0.66	0.28	0.67	0.65
Chlorite	-7.91	6.20	1.94	-5.74	-2.03	0.05	5.86	-6.79	4.70	1.42	2.73	-3.20	0.71	3.59
Fluorite	0.01	-2.61	-3.19	-0.53	-0.19	-0.20	-0.44	0.41	-2.42	-2.63	-1.67	0.21	-0.54	-1.26
Gibbsite	1.38	0.80	0.42	0.28	-0.83	0.37	-1.73	1.89	1.15	1.32	1.08	1.31	1.31	1.02
Goethite	7.04	7.16	6.85	6.79	5.38	7.02	6.09	7.36	7.59	8.80	7.71	7.85	7.77	7.54
Haematite	16.19	16.4	15.77	15.75	12.88	16.12	14.32	16.74	17.22	19.63	17.45	17.74	17.58	17.12
Illite	2.55	2.71	0.56	0.09	-1.70	0.23	-2.16	5.56	4.64	3.84	4.28	3.57	4.95	4.37
K-feldspar	0.73	0.85	-0.49	-0.35	-0.98	-0.70	-0.31	2.84	2.22	1.41	1.87	1.58	2.55	2.15
K-mica	9.23	8.16	6.05	6.04	3.15	5.77	2.03	12.25	10.16	9.71	9.67	9.85	10.81	9.82
Kaolinite	4.51	3.73	2.30	2.19	0.15	2.16	-1.38	6.61	5.22	5.01	5.16	4.83	5.61	5.04
Quartz	0.42	0.62	0.29	0.34	0.44	0.26	0.57	0.99	1.04	0.76	1.08	0.68	1.07	1.06
SiO ₂ (amorphous)	-0.72	-0.56	-0.88	-0.72	-0.66	-0.89	-0.51	-0.25	-0.19	-0.47	-0.16	-0.54	-0.15	-0.18

Table 5

Hydrogen and oxygen isotope compositions for sampled geothermal and nongeothermal spring waters.

	Location	$\delta^{18}O_{H2O}\%$	$\delta^2 H_{H2O} \ \%$
Hot water springs	Mahapelessa	-4.9	-28.0
	Rankihiriya	-6.5	-38.7
	Kanniyai	-5.5	-32.2
	Nelumwewa	-5.0	-29.9
	Mahaoya	-6.3	-37.4
	Marangala	-5.7	-35.0
	Kapurella	-5.9	-35.5
Cold groundwater	Mahapelessa	-6.2	-36.9
	Rankiriulpatha	-5.5	-32.8
	Kanniyai	-5.6	-33.5
	Nelumwewa	-5.1	-28.2
	Mahaoya	-5.5	-33.4
	Marangala	-5.5	-32.3
	Bibile	-5.9	-35.5



Fig. 5. $\delta^{18}O_{H2O}$ and $\delta^{2}H_{H2O}$ relationships (in % versus V-SMOW) for groundwater samples over local and GMWL-Global Meteoric Water Line (Rozanski et al., 1993). SW1 and SW2 are seawater samples that were taken from the shore and 3 km away from the shore, respectively.

therefore the data available in the ISOHIS database (http://isohis. iaea.org; accessed on 15 September 2011) were used to calculate the local meteoric water lines (LMWL) for the three different climatic zones of Sri Lanka (Table 6). Araguás-Araguás et al. (2000) also noted that stable isotope compositions of meteoric water is primarily affected by the amount of rainfall, which is a well-known feature of tropical oceanic islands such as Sri Lanka where seasonal variations of temperature are minimal. In these regions, the degree of isotope depletion of rainfall usually correlates well with the amount of precipitation. This was attributed to the different origins of moisture producing precipitation such as Indian Ocean and high altitude condensation (Jayasena et al., 2008). They also inferred that the isotopic composition of the precipitation in Sri Lanka is strongly influenced by the physiographical features as well as the local environmental conditions.

One of the main features of stable isotope analyses is that the isotopic signatures of geothermal and non-geothermal waters used in this study plot above the GMWL, defined by $\delta^2 H_{H2O} = 8.17 \delta^{18-1}$ O_{H20} + 10.35 (Rozanski et al., 1993) and close to the LMWLs drawn for the dry and intermediate climatic zones of Sri Lanka. This indicates that the recharge of all investigated subsurface waters mostly stems from local precipitation. With ocean waters having water isotope values of close to zero, this finding also reduces the possibility of admixture of ocean waters even though some springs are close to the shore and one could have expected a noticeable proportion of seawater input. Oxygen exchange processes between minerals and water in thermal springs are also not evident as both geothermal and non-geothermal springs show almost similar isotope signatures. Differences in mean isotope values between the geothermal springs are therefore rather related to discharge temperature and altitude of recharge zones.

Note that meteoric water collected from stations located in more inland areas of the dry and intermediate climatic zones are characterized by more depleted isotope compositions that are comparable to the isotope signatures obtained from hot- and cold-water samples. For instance, mean $\delta^{18}O_{H2O}$ values for Tanamalwila, Bathalagoda, Bandarawela, and Wannigama meteorological stations were -5.1%, -4.5%, -5.3%, and -4.5%, respectively. However, the Rankihiriya and Kanniya springs, which are located in the extreme north of the geothermal line, are characterized by more depleted $\delta^{18}O_{H2O}$ values (–6.2‰ and –6.5‰) compared to other thermal springs. This might be attributed to recharge at higher-elevations (Falcone et al., 2008). The Kantale IAEA meteorological station, which is located about 15 km away from the Kanniya hot water spring and about 35 km from the Rankihiriya hot water spring showed average $\delta^{18}O_{H2O}$ and $\delta^{2}H_{H2O}$ values of -4.2% and -18.0%. Again, this indicates that recharge has possibly taken place in a more inland and elevated region.

Table 6

Local meteoric water lines drawn for three climatic zones in Sri Lanka (data from IAEA-ISOHIS data base, http://isohis.iaea.org; accessed on 15 September 2011).

Climatic zone	δ ¹⁸ 0 ‰	δ ¹⁸ 0 ‰			Local meteoric water line (LMWL)
	Min	Max	Min	Max	
Wet zone	-11.8	-0.21	-72.7	5.1	$\delta^2 H = 7.77 \delta^{18} O + 10.62 (r^2 = 0.951)$
Dry zone	-9.83	2.17	-78.3	38.2	$\delta^2 H = 7.71 \delta^{18} O + 10.04 (r^2 = 0.950)$
Intermediate zone	-9.40	0.80	-54.0	-6.0	$\delta^2 \mathrm{H} = 7.63 \delta^{18} \mathrm{O} + 7.20 \; (r^2 = 0.960)$

5. Origin of the geothermal springs

The hydrogeochemical and isotope ($\delta^{18}O_{H2O}$, $\delta^{2}H_{H2O}$) characteristics of geothermal and non-geothermal waters from the Precambrian metamorphic terrains of Sri Lanka helped to narrow down the origin of the geothermal water. Various geothermometers were applied to explore deep reservoir temperatures. However, remarkable similarities of geochemical and isotope compositions of geothermal waters with those of the local non-geothermal waters indicates similar sources of both water types. Based on comparisons of $\delta^{18}O_{H2O}$ and $\delta^{2}H_{H2O}$ values with the local meteoric water lines, the origin of geothermal water recharge most likely stems from meteoric origin and circulates in crystalline rocks to different depths. In addition, close scattering of data around local meteoric water lines indicates that precipitation of the dry and intermediate zones recharges the springs without significant evaporation prior to the infiltration. This indicates that infiltration of precipitation is fast and does not allow evaporative alteration of the isotope composition of the waters. Furthermore, differences of water isotopes between geothermal waters could be related to the mean recharge elevations of thermal groundwater. All springs that are located below or close to the 100 m contour line and have more negative isotope values than the weighted averages of the local meteoric water lines indicate that recharge possibly occurs at higher altitude locations either in the dry zone and/or in the intermediate climatic zone.

Geothermometric calculations based on the dissolved silicaquartz/chalcedony regressions indicated that geothermal springs in Sri Lanka have shown low enthalpies in which the highest reservoir temperatures were recorded at Marangala and Nelumwewa springs. It also indicates a significant degree of heterogeneity in the subsurface heat distribution where the highest temperatures may prevail in the middle of the geothermal zone. Note however, that the calculated temperatures may deviate from real temperatures because the necessary geochemical equilibria that are a fundamental assumption for these results may not have been reached. This is evident from isotope and geochemical similarities between shallow groundwaters and thermal springs and may suggest a fast circulating system. Knowledge about formation depths of the thermal springs are difficult to establish. However, in general the average geothermal gradient in many parts of the world is 25-30 °C/km and with that at least 3 km depth are required to reach temperatures of up to 90-100 °C (Fridleifsson et al., 2008).

The geothermal waters from Sri Lanka likely stem from much shallower depths because fast conduit circulation systems are unlikely reach down to depths of several thousand meters. This would argue for steeper geothermal gradients. This is also confirmed by the geochemical characteristics. The steeper thermal gradient may be related to the residual heat along the Highland– Vijayan boundary zone, which has been recognized as a possible Precambrian tectonic or thrust zone (Dissanayake, 1985; Kröner et al., 1991).

The geothermal water from the metamorphic terrains studied here also indicates restricted rock–water interactions and with this more shallow circulation pathways. For instance, less dissolved minerals in geothermal waters suggests that waters rise to the surface without equilibration with aquifer rocks. This is particularly possible in hard rock terrains that usually have more difficulty to weather minerals. The restricted dissolution of quartz or amorphous silica due to shorter equilibration times also compromises the use of soluble mineral based geothermometers.

The stable isotope result of the subsurface waters and their similarity to the local meteoric water lines indicate that sea water intrusion must play a negligible role. Moreover, possible steam condensation or heating or evaporation may also have affected the original isotopic composition of meteoric waters. However, this is not the case in hot springs in Sri Lanka because such processes would impart isotope changes of the waters sampled that in turn would drive away the samples from the meteoric water line. Since all samples are close to the LMWL any significant evaporation, formation of steam and condensation did very likely not occur.

The most plausible mechanism is that meteoric waters readily percolate to shallow depths along structurally weak zones in the high-grade rocks, are heated and then rise to the surface through fault or fracture conduit zones at a reasonable pressure. Areas of faults crossing are more favorable to form warm springs (Fouillac and Michard, 1981; Kharaka and Mariner, 2005). In this manner they create a line of hot springs in the Precambrian Crust.

6. Conclusions

Analyses and comparison of hydrogeochemical and isotopic characteristics of waters from thermal springs and non-thermal groundwater collected close to thermal springs reveal that both waters are hydrogeochemically and isotopically similar in composition. Stable Isotope data suggest that the origin of both thermal and non-geothermal water recharge is almost all exclusively precipitation. Low concentrations of trace elements in both type of water also provide evidences for limited rock-water interactions hence limited deep circulations. The recharge elevations are consistent with thermal groundwater having been recharged in the wet dry zone, as part of the regional flow system in the basin. The best suitable mechanism of formation is that infiltrating water percolates downwards through faults and fractures, and is heated by a steeper geothermal gradient. It then rises to the surface along permeable zones. The non-geothermal waters also circulate in the same way, but percolate into much shallower depths. Further investigations including radiogenic and stable isotope measurements at higher frequencies as well as geophysical testing would help to fully characterize this geothermal system in more detail.

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