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GEOCHEMICAL CHARACTERISATION OF WATERS AND GASES OF ISCHIA ISLAND (ITALY).

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Key Words*:* gas and water geothermometers, fluid geochemistry, Ischia Island, $\delta^{13}C_{CO2}$, gas-water-deep fluids interactions.

ABSTRACT

Between 1994 and 1995 gas samples from some fumaroles and thermal waters were collected on Ischia Island. The chemical composition of the shallow and deep natural hydrothermal fluids discharged is related to the main hydrological and lithological characteristics of the rock formations present in the reservoir. The calculated mass vapor fraction (y) of the reservoir is always positive suggesting the presence of a biphase system where the dominant liquid has a temperature ranging from 240 to 280°C. The liquid phase is made up of modified seawater, heated fresh seawater and meteoric water. The mixing between these components together with interaction processes with the surrounding rocks is the origin of all of the islands groundwaters. The gaseous phase is mainly carbon dioxide with variable contents of helium that reflect different degrees of interaction with shallow groundwaters. The isotopic composition of the helium and the CO_2 of these gases is characteristic of fluids that originate in a mantle that has undergone crustal contamination. The helium values are from 1 to 3 R/Ra and values of $\delta^{13}CO_2$ among -5 and –2 δ‰. The negative $\delta^{13}C$ value with respect to typical magmas in the Mediterranean (with $\delta^{13}CO_2$ ranging from 0 to -2δ‰) is the result of deep gases and groundwater interaction during their rise towards surface. The equilibrium temperature of the deep reservoir, found by using the chemical composition of the sampled gases, is uniform at 280°± 20°C. The homogeneity of the equilibrium temperatures of the sampled gases in different areas of the island probably indicates the existence of a common geothermal reservoir from which the gases originate. The differences in the equilibrium temperatures, estimated from the water's chemical composition, are due to mixing processes between modified sea water that represents the deep recharge of the system, and shallow groundwaters recharged by meteoric and marine waters at lower temperatures. The amount of steam in the reservoir is below 2% of total water content. The calculated values of pCO_2 in the reservoir are highly variable and range between 1 and 28 bars. The observed values do not show any correlation with other parameters and do not allow the evaluation of pCO_2 in the geothermal reservoir.

1. INTRODUCTION

Ischia, Phlegrean Fields and Vesuvius are part of the classic active volcanic area of the alkali-potassic Quaternary Roman Province of central-southern Italy developed within the Campanian graben, on the central-western margin of the apennine chain. The volcanism is thought to be a consequence of tensional tectonics with NW-SE and NE-SW trends induced by the opening of the Tyrrhenian sea basin. This volcanic activity started about 2.0 Ma ago and has lasted until the present day with the historical eruption of Vesuvius, Phlegrean Fields and Ischia. The NE-SW tectonic trend marks the alignment of the volcanic areas of Phlegrean Fields and Ischia that were formed by many eruptive centers and are characterized by the presence of many thermal springs, fumaroles, frequent earthquakes and bradyseismic phenomena (Santacroce, 1987, Vezzoli, 1988). The island of Ischia lies on the northwest edge (rim) of the Bay of Naples, opposite Capo Miseno. Oblong in shape, Ischia has a circumference of 34 km and a surface area of 46 km^2. The island is part of a volcanic field that was larger that the present island. Mt. Epomeo, an active volcano-tectonic horst raised by the intrusion of a superficial magmatic chamber, constitutes the highest part of the island (787 m a.s.l.) and the only part involved in the tectonic uplift (Vezzoli, 1988). The last eruption on the island took place in 1301 A.D. at Arso. There have been destructive earthquakes more recentely, the last in 1833 when the entire town of Casamicciola was destroyed. Geothermal research was carried out on the island in the 1940's (Penta and Conforto, 1951). Several wells were drilled there and the hottest temperature (232°C) was measured in the deepest well at 1150 m. Many thermal springs (T ~90°C) and fumaroles (T~100°C), and shallow earthquakes (the last one in 1881, Mercalli, 1884) testify to the persistent state of activity of the magmatic system. The aim of this work is to evaluate the thermodynamic features (P, T and vapour fraction) of the island's deep reservoir feeding shallow fluids.

2. HYDROGEOCHEMICAL FRAMEWORK

Between 1994 and 1995 gas samples from some fumaroles and thermal waters were collected on Ischia Island. Figure 1 shows the location of sampled fluids. The chemical composition of water samples from Ischia samples (Table 1) suggests that these are the result of the mixing of seawater and low salinity waters of probable meteoric origin, as hypothesised by De Gennaro et al., (1984), and Panichi et al., (1992). Water-rock interaction processes are particularly evident in many samples. The chemical composition of Ischia thermal waters are due to the mixing of at least three end-members: a "marine" modified component (e.g. S. Lorenzo sample, the most saline, with the highest concentration of B and t=90°); a "fresh" meteoric component that has undergone little rock interaction (e.g. sample 11, the least saline, with the lowest concentration of B, and t=28°C); a "modified" meteoric component due to rock interaction processes (e.g. samples 15 and 18, intermediate salinity and B concentration and t=30 and 53°C respectively). Sample 5 (S. Lorenzo), represents waters of marine origin, subsequently modified and heated. In fact, the salinity of these water samples reaches values sometimes higher then that of seawater. The distribution of sample data points in the ternary diagram in Figure 2 (Giggenbach, 1988) shows that most of the waters fall within the fields of non-equilibrium or partial equilibrium with the rocks. Samples 11 and 5 identified as mixing end members, fall within the fields of immature and partially mature waters, respectively. The intermediate samples tend to lie in positions of increasing maturity towards the saline end member (5). Samples 14, 15 and 16 fall within the field of full equilibrium with equilibrium temperatures between 100 and 160° C. Samples 5, 8 and 18 show Na/K ratios typical of high temperature equilibrium (240 to 280°C) in agreement with temperatures measured in the perforation wells drilled by SAFEN in the 1950's (Penta and Conforto, 1951). From the application of various geothermometers in mature water samples an estimation of the deep reservoir waters was made.

Table 2 shows the calculated temperatures of the more representative samples. The data clearly shows that the Na/K, Na-K-Ca and Na-Li geothermometer ratios have comparable temperatures between them (even though Na-Li of Fouillac always shows T lower than that of Kharaka), while the SiO_2 and Mg ratios show a lower T. Water-rock exchange reactions involving Mg proceed relatively fast at low temperatures. This geothermometer, like that of $K/Mg^{1/2}$, appears to be a good indicator of the last temperature of water-rock equilibration in ascending water. A contributing factor in this apparent re-equilibrium is that both Mg and Li are minor constituents in most thermal waters. Thus reactions involving a relatively small amount of these constituents may have a great effect on their ratios. When the estimated temperatures using $K/Mg^{1/2}$, $Li/Mg^{1/2}$and other geothermometers (particularly Na-K-Ca and silica) are in agreement, it is fairly certain that water-rock equilibration at the estimate temperature range was reached and that water-rock reaction during surface flow was negligible. Agreement between $K/Mg^{1/2}$ and $Li/Mg^{1/2}$ geothermometer temperatures, but higher estimated temperatures when using other geothermometers, may indicate a relatively short time in the reservoir, and intermediate temperatures where water-rock equilibrium only occurs with respect to the most reactive phases involving Mg. The estimated temperatures for the waters from the San Lorenzo well, ranging from 247 to 274°C (Table 2) confirm the hypothesis that this is the most representative sample of the deep reservoir that feeds the thermal manifestations of the island. Furthermore, the saturation index for this well (SI=logAP/Kps) for various mineralogical species at various temperatures, using microcline to balance the Al and SiO_2 in equilibrium with quartz was taken. Considering the uncertainty of ±0.25, the range of equilibrium temperature shown in the SI vs T diagram (Fig. 3) varies within the 260 and 300°C range. The average temperature of the reservoir can be considered near 280°C. This value is compatible with that measured with the geothermometers in a liquid phase.

3. GAS COMPOSITION

Free gas samples of Ischia are essentially CO_2-dominant gases mixed with air in various proportions (Table 3). In the He-CO_2-N_2 plot (Fig. 4) the samples show wide variations in the He/CO_2 ratio that reflect different degrees of interaction between fluids of deep and shallow origin. This variation is a consequence of the difference in solubility of He and CO_2 in groundwater and the chemical-physical conditions of the groundwater (T, pH, TDS). The free gas samples cluster near the CO_2 vertex and follow two distinct trends (Fig. 4): the first with an increase in the He/CO_2 ratio due to interaction processes with the groundwater; the second with an increase in the N_2/CO_2 ratio caused by atmospheric contamination. A possible explanation for the high concentrations of He in fumaroles 28, 29, 32 and 33, is that CO_2–rich fluids rose from depth with He/CO_2 ratios corresponding to that the gas phase dissolved in magma, and became enriched in He after interaction with shallow, CO_2 –unsaturated groundwaters. The extent of this reaction is a function of different He and CO_2 solubility coefficients (He=8.7 ml/l and CO_2 = 759 ml/l at STP condition), as well as the pH of the groundwater, since CO_2 solubility depends on this parameter (Capasso et al., 1996, 1997). Local groundwaters are able to dissolve large amount of CO_2 thus enriching He in the residual phase. On the other hand, the dissolved gases show a He/N_2 ratio (solubility coefficients of N_2 =14.6 ml/l at STP condition) that varies to atmospheric probably due to interaction with shallow air saturated waters. Moreover, the alignment of these samples toward the CO_2 vertex in Fig. 4 indicates different degrees of interaction with deep fluids rich in CO_2. Starting from the method developed by Giggenbach (1980) for New Zealand hot water systems, D'Amore et al. (1982) and D'Amore and Celati (1983) showed that combining gas solubility and the equilibrium constants of two reactions, both the temperature and the mass vapour fraction (y) of the reservoir can be calculated. The calculated mass vapour fraction (y) of the reservoir is always positive (Table 4), hence suggesting the presence of a biphase system.The amount of steam in the reservoir is below 2% of total water content. The calculated values of pCO_2 in the reservoir are highly variable and range between 1 and 28 bars. The observed values do not show any correlation with other parameters and do not allow the evaluation of pCO_2 in the geothermal reservoir. The equilibrium temperatures of fumarole gases were estimated using six different geothermometers in a gaseous phase reported in appendix 1 (D'Amore & Panichi 1980; Arnorsson & Gunnlaugsson, 1985; Arnorsson et al., 1983; Giggenbach et al., 1980; Saracco & D'Amore 1989; Giggenbach, 1987). The temperatures obtained are listed in Table 4 along with the vapour fraction y and the pCO_2. The information provided by the six gas-geothermometers was used to calculate the equilibrium temperature sufficiently consistent between them. The average temperature calculated considering all gas-geothermometers and all sites (n=27) is 288° C ± 27. Moreover, by using the graph method proposed by Giggenbach (1991) based on CO/CO_2 and CH_4/CO_2 ratios (Fig. 5), we were able to estimate the temperature of the deep reservoir that feeds this volcanic system. The resulting temperatures ranged from 270 to 330°C. These results are consistent with the estimated values of the cationic geothermometers, calculated by the saturation index for sample 5 and with the temperature of the ISFV2 well drilled in the 1950's (Penta & Conforto, 1951). Other authors have also estimated the deep equilibrium temperatures of the Ischia gases, Carapezza et al. (1988) obtained values ranging from 214 to 364 °C for 9 different sites, in agreement with our estimates. The temperatures calculated by Panichi et al. 1992 ranged from below 200 to above 250°C, slightly lower than our calculated values. Tedesco (1996) also calculated the deep temperatures on the basis of chemical and isotopic equilibrium of carbon species. The obtained values ranged from 280 to 550°C. The highest T values were also supported by temperatures calculated by isotopic reactions between CH_4 and CO_2. However, such high values are probably due to different degrees of contamination from shallow CH_4-rich hydrothermal fluids. In this case the isotopic values of $\delta^{13}C$ of CH_4 reported by Tedesco (1996) do not allow discrimination of the origin of methane.

4. GAS ISOTOPE COMPOSITION

He and C isotopic compositions give useful information about fluid origin and about the chemical and isotopic processes that gases undergo as they rise towards the surface. In particular, the He isotopic composition is utilised to determine the different origins of fluids because of its low reactivity and solubility in the waters. CO_2, on the other hand, is highly reactive and soluble in waters and it is involved in several chemical and isotopic equilibria that modify the original composition of fluids. The $\delta^{13}C_{CO2}$ and $^3He/^4He$ of the gases of Ischia are given in Figure 6 together with those of MORB, and other active volcanic areas in the Mediterranean Sea (Etna, Vulcano, Plegrean Fieds). Like the other volcanic areas, helium and $\delta^{13}C_{CO2}$ values of Ischia do not fall into the MORB compositional field (Fig. 6). Helium isotopic ratios of Ischia fluids show values ranging from 1 to 3.6 Ra ($Ra= 1.4*10^{-6}$), very similar to those of Phlegrean Fields, and clearly lower than those in fluids from Etna and Vulcano. These values seem to indicate that gases originated in a mantle that locally underwent crustal contamination. In particular, low $^3He/^4He$ values can be related to the particular abundance of U and Th values in the host rocks that release particles of α (radiogenic ^4He). $\delta^{13}C_{CO2}$ values show a wide range from 0 to –5 ‰ vs PDB (Table 3), similar to those observed in other volcanic active areas in Italy (ranging from -3 to 0‰). Such values are more positive than those related to MORB (-8 to -5 ‰; Taylor et al., 1967, Deines, 1970). The more positive isotopic values of these samples with respect to MORB, rather than reflecting shallow contamination processes by CO_2 derived from thermal metamorphism of carbonate sediments of the basement of the volcano, would suggest that the source of magmatic CO_2 is already contaminated (Marty et al. 1994, Hoernle et al., 1995). Such a hypothesis is soundly supported by the results obtained from recent geochemical studies in the Mediterranean area (Capasso et al., 1996, 1997, 1998, 1999; Giammanco and Inguaggiato, 1996; Inguaggiato and Italiano, 1998; Inguaggiato and Pecoraino, 1998; Giammanco et al., 1998).

5. SUMMARY AND CONCLUSIONS

Geochemical studies of the thermal waters and gases of the Ischia Island have permitted to identify and characterise deep and surface components that feed the hydrothermal system. Deep magmatic fluids are characterised by CO_2-rich gases with helium isotopic values ranging from 1 to 3.6 R/Ra and $\delta^{13}C$ of CO_2 values around –2 δ‰. These gases have an isotopic composition reflecting crustal contamination of the mantle. In particular, low $^3He/^4He$ values can be related to the particular abundance of U and Th in the host rocks that release radiogenic ^4He. The deep reservoir has a liquid-dominant biphase system ($y \cong 0.02$) with equilibrium temperatures near 280°C and PCO_2 up to 30 atm. The liquid phase is made up of modified seawater that is enriched in B, K, Na and depleted in Mg because of intense processes of exchange with the rocks. The equilibrium temperature of the deep reservoir, determined using the chemical composition of the sampled gases, is uniform and varies between 280°± 20°C. The homogeneity of the equilibrium temperatures of the sampled gases in different areas of the island probably indicates the existence of a common geothermal reservoir from which the gases originate. The differences in the equilibrium temperatures, estimated from the water's chemical composition, are due to the mixing processes between modified sea water that represents the deep recharge of the system, and shallow groundwaters recharged by meteoric and marine waters at lower temperatures. Concluding, the chemical and isotopic differences observed in Ischia Island fluids are linked to interaction processes (equilibria and/or kinetics) between deep and shallow fluids. For this reason we exclude the existence of different reservoirs feeding the geothermal system like Tedesco (1996) proposed.

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Table 1. Chemical composition of thermal waters (selected data). Contents are expressed in mg/kg.

n.	data	t	pH	SiO_2	Na	K	Ca	Mg	Li	Cl	SO_4	Alc	B
4	ott-95	48	6.5	100	10744	663	651	1305	n.d.	20235	2329	1586	6.3
5	gen-94	73	5.7	170	8253	1341	300	277	41	14633	703	229	n.d.
5	ott-95	66	6.0	180	14058	2031	436	395	n.d.	23855	1307	275	16.9
8	ott-95	47	7.7	150	1125	143	16	4	n.d.	465	572	1708	2.3
11	ott-95	28	6.4	0	166	25	141	17	n.d.	79	179	580	0.9
15	ott-95	30	6.9	n.d.	1426	12	33	2	n.d.	378	693	2440	2.3
16	ott-95	90	7.0	n.d.	3635	112	88	9	n.d.	5037	451	531	8.5
18	ott-95	53	8.5	n.d.	959	142	2	0	n.d.	333	334	1642	2.3
21	ott-95	34	5.5	n.d.	11678	569	521	1310	n.d.	19978	2494	528	6.8

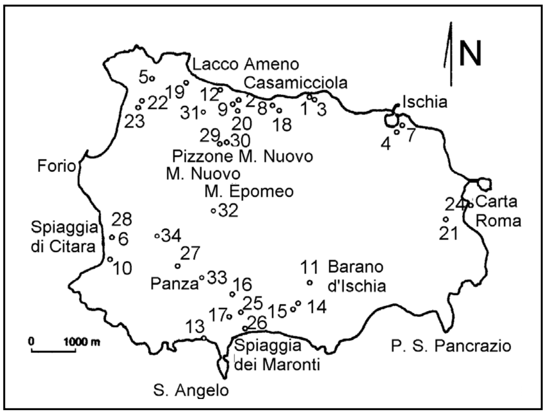

Figure 1. Location map of sampled fluids

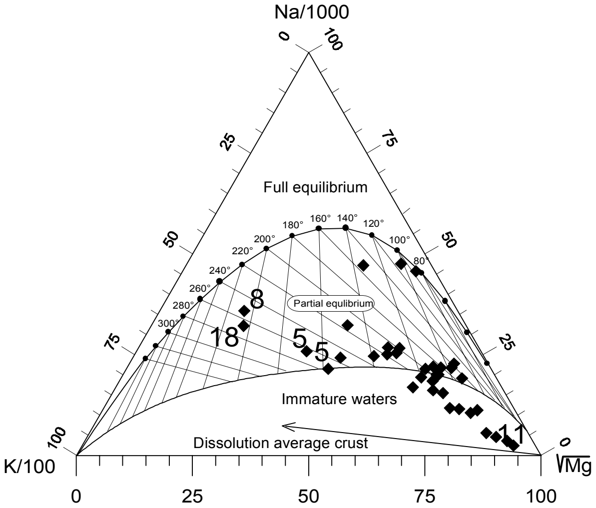

Figure 2. Relative Na, K and Mg contents of thermal waters. Isotherms calculated by use of Na-K and K-Mg geothermometers are given. Full and partial equilibrium and immature waters are also shown.

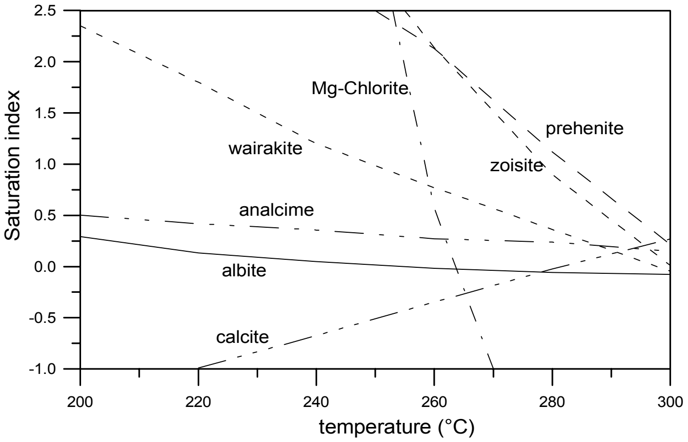

Figure 3. Temperature (°C) estimated for n. 5 well by using saturation indexes for various mineralogical species at temperature ranging from 200 to 300°C.

Table 2: Calculated temperatures obtained by applying cationic geothermometers (1= Truesdell & Fourier, 1975; 2: Tonani, 1980; 3: Arnorsson et al., 1983; 4: Fournier, 1979; 5: Nieva and Nieva 1987; 6: Giggenbach, 1980; 7: Kharaka and Mariner, 1989; 8: Fouillac and Michard, 1981; 9: Fournier, 1973)

Samples	t Na/K^1	t Na/K^2	t Na/K^3	t Na/K^4	t Na/K^5	t Na/K^6	t Na-K-Ca^7	t K/Mg^8	t Li/Mg^9	t Na/Li^7	t Na/Li^8	t SiO_2^{9}
Thermal C.1	139	143	148	177	164	194	197	110	75	114	73	137
S.Lorenzo	247	258	251	263	248	274	266	160	160	242	217	169
P. SAFEN	168	173	175	200	187	216	215	134	128	187	152	183
H. Reginella	167	173	175	200	187	216	200	127	118	168	131	153

Table 3. Chemical and isotopic composition of gas samples. He, H_2, CO, CH_4 contents are expressed in ppm, O_2, N_2 and CO_2 contents in %vol. Isotopic carbon values of CO_2 are expressed in δ‰ vs. PBD.

n.	Samples	Data	t	He	H_2	O_2	N_2	CO	CH_4	CO_2	$\delta^{13}C_{(CO2)}$	$^3He/^4He$
24	Carta Romana	Jan-94	15	2.0	<2	0.4	1.8	<1	53.5	95.8	-2.05	n.d.
24	Carta Romana	oct-95	16	2.0	<2	0.4	1.0	1.9	0.7	96.4	-2.49	0.93
25	Terme Linda	Jan-94	98	7.9	101	2.1	7.5	<1	75.1	88.9	-2.97	n.d.
25	Terme Linda	oct-95	98	21.0	121	0.6	2.1	2.6	75.3	95.0	-3.01	1.63
26	Spiaggia Maronti	Jan-94	103	5.4	2.7	1.5	4.4	3.8	49.0	96.5	-3.04	n.d.
26	Spiaggia Maronti	oct-95	101	13.9	558	3.2	10.1	10.6	45.0	88.3	-3.26	3.69
27	Panza C.Sportivo	oct-95	98	9.3	174	6.5	25.5	12.3	24.7	76.6	-3.52	n.d.
28	Pozzo Safen Citara	Jan-94	50	23.0	1416	0.2	3.6	<1	36.7	97.1	-4.49	n.d.
29	Pizzone Monte Nuovo	oct-95	99	113.0	11800	0.4	1.7	2.3	24.8	89.5	-5.00	3.56

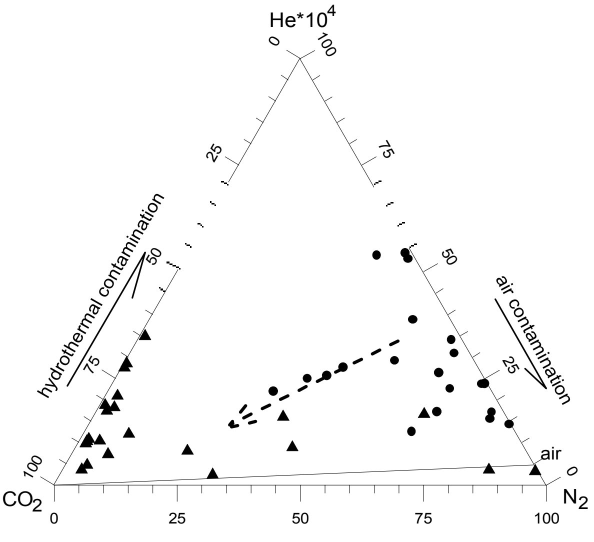

Figure 4. CO_2, N_2 and He*10^4 contents of fumarolic gases. External arrows indicate effects of hydrothermal and air contamination, dashed arrow indicates the effect of interaction between local groundwaters and deep gases. Full triangles are deep gases, full circles are dissolved gases.

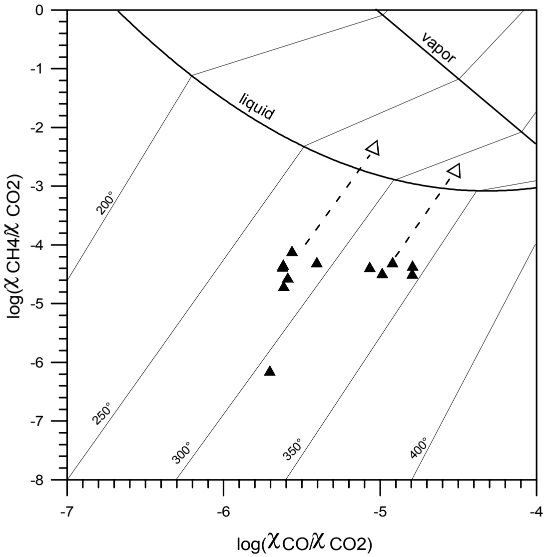

Figure 5. Evaluation of temperature (°C) of the Ischia geothermal reservoir using CH4, CO, CO2 contents of discharged gases.

Table 4. Calculated temperatures obtained by applying gas geothermometers (1: D'Amore and Panichi, 1980; 2: Arnorsson and Gunnlaugsson, 1985; 3: Giggenbach, 1987; 4: Saracco and D'Amore, 1989; 5: Giggenbach, 1980).

Samples	t^1	t^2	t^2	t^3	t^4	t^5	y^4	$pCO_2{}^4$
Pizzone Mt Nuovo	283	288	313	281	270	n.d.	0.018	0.94
Panza C.Sportivo	340	271	273	320	330	n.d.	0.025	17
Mt Citto	313	297	313	300	300	328	0.018	28
Mt Corvo	226	268	303	285	300	231	0.002	4
Cimmento Rosso	266	277	281	280	280	271	0.005	5

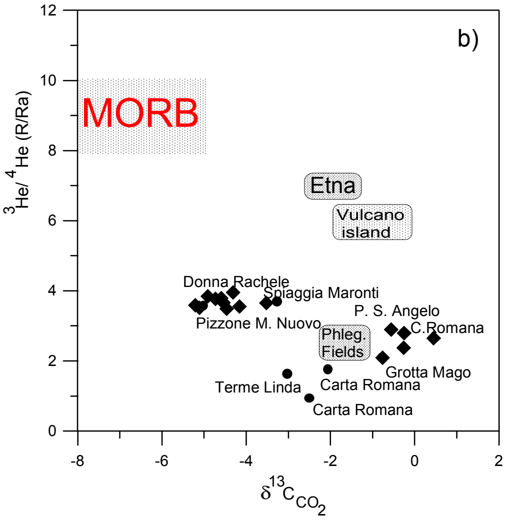

Figure 6. The $^3He/^4He$ vs $\delta^{13}C_{CO2}$ for Ischia gases.