



Research in Shallow Marine and Fresh Water Systems

Detection of fresh and thermal waters over an island with extinct volcanism: the island of Salina (Aeolian arc, Italy)

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Abstract

A geochemical prospection of the fluids circulating over a volcanic island highlighted the existence of reservoirs of both fresh and thermal water although only a few natural manifestation, mainly occurring in submarine environment, provide clues of their existence. Coupling the information provided by the diffuse gas bubbling and the low-salinity waters discharged from the sea floor off the Island with those from the low-salinity and thermal waters collected from in-land wells drilled in recent times we found that a large amount of fresh and thermal waters are continuously discharged at the sea level. The thermal waters, collected at a temperature of 32-36°C, show a typical marine recharge with equilibrium temperatures estimated to be in the range of 120-160°C. As matter of fact, the small island holds relatively large reservoirs of both fresh and thermal waters. The geochemical features of the circulating fluids are driven by water-rock and gas-water interaction processes, while the heat supplied to the geothermal reservoir comes from volcanic rocks that are still cooling down after the end of the last volcanic activity supposed to be not older than 13ky.

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1. Introduction

Small islands are often very well known tourist places, exploited for tourist purposes because of their valuable natural environment. Small islands located far away from the main land, however, have normally two main limitations to solve: electrical power and fresh water.

As an example of such a situation a volcanic island belonging to the Aeolian arc (Southern Tyrrhenian sea), about 16 miles off the northern coasts of Sicily (Italy) has been considered for the development of a research project aimed to recover geochemical information to better understand the origin and the role of the circulating fluids and their relationships with its natural volcanic environment.

The island of Salina is the second largest island of the volcanic Aeolian archipelago (Southern Tyrrhenian Sea) and includes five main subaerial eruptive centres ranging in age from 430 to less than 13 Ky distinguished in two main cycles of volcanic activity [1, 2]. The younger cycle (100-13 Ka) built up the andesitic cone of Monte dei Porri (Figure 1) and ended with the formation

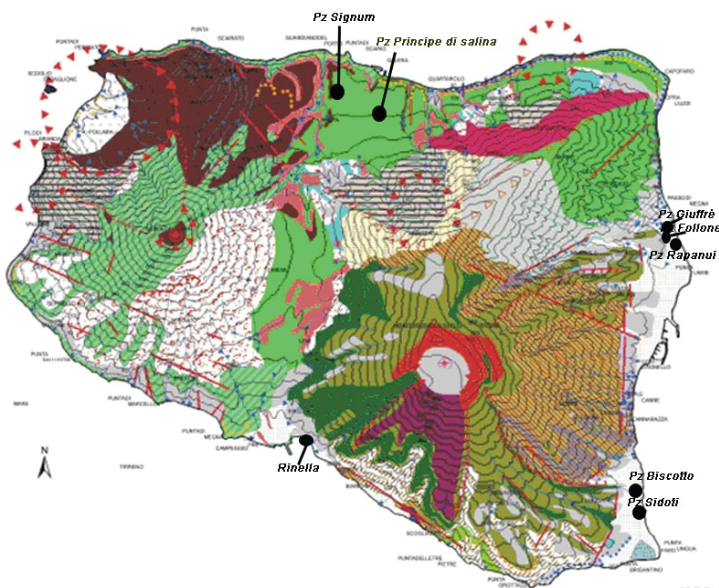


Figure 1 – Geological map of the island of Salina and location of the sampling sites

of the explosion crater of Pollara (Fig. 1) on the northwestern edge of the island [2, 3]. The volcanic activity resumed at least three times during the last 30 Ky within the Pollara depression with the effusion of the andesitic to dacitic lava flows of Punta di Perciato (30 Ka) followed by two explosive eruptions (Lower and Upper Pollara pyroclasts, [1]).

As a consequence of the cooling of the old magmas, volcanic fluids are released and a persistent gas bubbling was detected off all the coasts [1]. The released gases are CO₂-dominated and marked by a helium isotopic ratio in the range of 5.5-6Ra denoting the magmatic origin of the gas phase. In land a thermal spring was known in past (nowadays disappeared) and apart from an old well excavated at the SE corner of the island, there are no other evidences of cold springs although the island is covered by a luxuriant vegetation.

In recent times (over the last 20 years), some wells were drilled on the eastern coast at shallow depth (3-10m), to recover low salinity water for local concrete manufactures, besides two deep wells drilled on the northern coast to recover thermal waters. The shallow wells on the eastern coast supply cold waters with a salinity well below the sea-water; the two deep wells are fed thermal waters ($T=32-38^{\circ}\text{C}$) exploited for local spas.

Table 1. Proper names, geographical coordinates and field data of the sampled wells. Geographical coordinates in UTM WGS84. See figure 1 for their location on the Island. (E.C. = Electrical conductivity in mS/cm; TDS in g/l)

Site	Name	Latitude	Longitude	T°C	pH	E.C.	TDS
Well 1	Giuffrè	488884	4268697	19.7 – 21.0	8.2	5.65	6.8
Well 2	Biscotto	488590	4266029	19.2	7.2	14.33	8.5
Well 3	Follone	488885	4268644	19.0 - 21.3	7.5	2.46	3.6
Well 4	Lo Schiavo	488916	4268512	17.2 – 21.0	7.2	11.46	1.2
Well 5	Rapanui	488916	4268512	18.5 – 22.4	7.5	0.87	5.4
Well 6	Sidoti	488424	4265685	16.0 – 20.5	8.2	7.01	4.1
Well 7	Principe di Salina	485910	4269862	36.0 – 39.0	6.0	69.40	39.6
Well 8	Signum	485532	4270118	25.0 – 31.5	5.3	24.25	28.0

In the mainframe of an INGV-DPC project, a geochemical survey was carried out to improve our knowledge on the presence and distribution of the natural water resources of the Island, starting from the available former information [4]. A repeated mid-term water sampling was carried out with the aim to gain an insight into the geochemical features of the fluids circulating over the Island (including waters and gases as dissolved gas phase) besides possible temporal changes.

The collected results allowed us to recognize the contemporary presence of high-temperature geothermal fluids and cold, fresh, waters which geochemical features mark the occurrence of Gas-Water (GWI) and Water-Rock Interaction (WRI) processes responsible for the chemical and isotopic composition of the waters and the dissolved gases.

2. Sampling and analytical methods

A total of 33 water samples have been collected from 8 different boreholes (see Figure 1 for their geographical location) over a time span of 19 months. The waters discharged by 6 shallow depth wells displayed an outlet temperature ranging from 16 to 22.4°C and hereafter are conventionally defined as “cold”. Contrastingly, the deep wells (2) drained thermal waters with a temperature in the range of 30-39°C.

The water samples were collected in triplicate and stored as “as is sample”, filtered sample (0.45 μm filter) and “filtered and acidified” (HNO_3 Suprapur-grade acid). All the water samples were analyzed for their chemical and isotopic composition. The analytical determinations included field measurements ($T^{\circ}\text{C}$, pH, Eh and electrical conductivity, EC) and laboratory analyses. On the field, the temperature was measured by mercury thermometer (resolution 0.01°C; error $\pm 10\%$) while pH, Eh and EC were measured by electronic instruments calibrated in situ using buffer solutions. In the laboratory, the chemical composition was determined by liquid chromatography (Dionex 2001) using a Dionex CS-12 and a Dionex AS4A-SC column for cations and anions determinations, respectively. The HCO_3 content was measured by standard titration procedures with hydrochloric acid.

Isotope determinations (D/H and $^{18}\text{O}/^{16}\text{O}$) on water samples were performed by equilibration technique ([5] for oxygen) and water reduction (hydrogen production by using granular Zn, [6]) respectively. Measurements were carried out using a Finnigan Delta Plus mass spectrometer (Hydrogen) and an automatic preparation system coupled with an AP 2003 IRMS (Oxygen). Analytical precision for each measurement is better than 0.2 ‰ for $\delta^{18}\text{O}$ and 1 ‰ for δD .

The dissolved gases were extracted from water samples stored in 240ml glass bottles sealed in the field by silicon/rubber septa using special pliers. The extracted gases were analyzed for their chemical composition and for the isotopic composition of helium and total dissolved carbon (TDC). The chemical analyses were carried out on the gas phase extracted in the laboratory after the attainment of the equilibrium (at constant temperature) between the water sample and a known volume of host, high purity gas (argon), injected inside the sampling bottle (see [7, 8] for details).

The analyses were carried out by a gas-chromatograph (Perkin Elmer 8500) equipped with a double detector (TCD-FID) using argon as carrier gas.

Helium isotope analyses were carried out on gas fractions extracted following the same procedure as for the gas-chromatography. The samples were purified following already proposed procedures (e.g. [9, 10]). The isotopic analyses of the purified helium fraction were performed by a static vacuum mass spectrometer (GVI5400TFT) that allows the simultaneous detection of ^3He and ^4He -ion beams, thereby keeping the $^3\text{He}/^4\text{He}$ error of measurement to very low values. Typical uncertainties in the range of low- ^3He (radiogenic) samples are within $\pm 5\%$.

The $\delta^{13}\text{C}_{\text{TDC}}$ (TDC = total dissolved carbon) was measured following a method based on the chemical and physical stripping of CO_2 . The $\delta^{13}\text{C}$ isotopic values were measured using a Finnigan Delta Plus mass spectrometer and the results expressed in ‰ vs. V-PDB standard. The standard deviation of $^{13}\text{C}/^{12}\text{C}$ ratio is ± 0.2 ‰.

3. Results and discussion

The location of the sampled boreholes is given in Table 1 as UTM-WGS84 coordinates. The chemical composition of the sampled waters, their stable isotopes composition (δD , $\delta^{18}\text{O}$ and $\delta^{13}\text{C}_{\text{TDC}}$), composition of the dissolved gases are shown in figures 2-8 where the local sea water is shown as reference. The geochemical features are given in table 4 together with the $^3\text{He}/^4\text{He}$ and $^4\text{He}/^{20}\text{Ne}$ ratios.

3.1 The cold and thermal waters

The analytical results show the presence of two main group of waters (Table 1): wells with outlet temperatures ranging from 16 and 21°C, TDS values $<7\text{g/l}$ and wells with temperature $>30^\circ\text{C}$ and TDS $>25\text{g/l}$.

The cold waters display low HCO_3 contents (normally well below 10meq/l) in contrast with the thermal waters which bicarbonates content is always above 20meq/l besides Cl and Na concentration similar to the local sea-water.

As Br is typically enriched in sea water to the respect of the continental waters and groundwaters, the presence of a marine component in the sampled waters can be shown by a Cl-Br diagram. Figure 2 shows how the samples fall on a mixing line between two components: a low-salinity end member and the sea water, showing that the cold waters are fresh-type waters contaminated by marine waters at low extents, while the thermal waters are mainly composed by seawater.

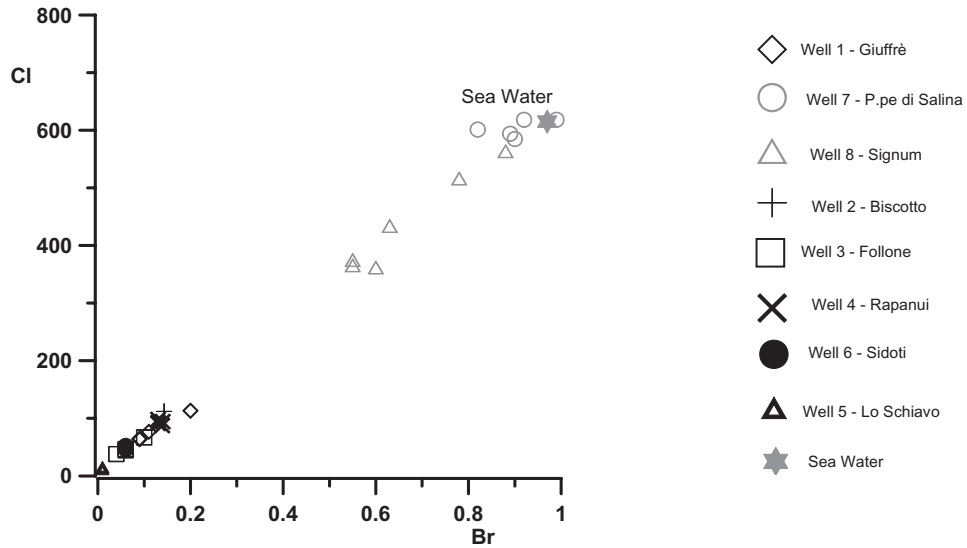


Figure 2 – Cl vs Br content of cold and thermal waters. Black symbols identify the cold waters; grey symbols for thermal samples. Sea water data are reported as a reference, however sea water is one end-member together with the low salinity water of well 4. The sample distribution denotes variable mixing proportions between the two end members

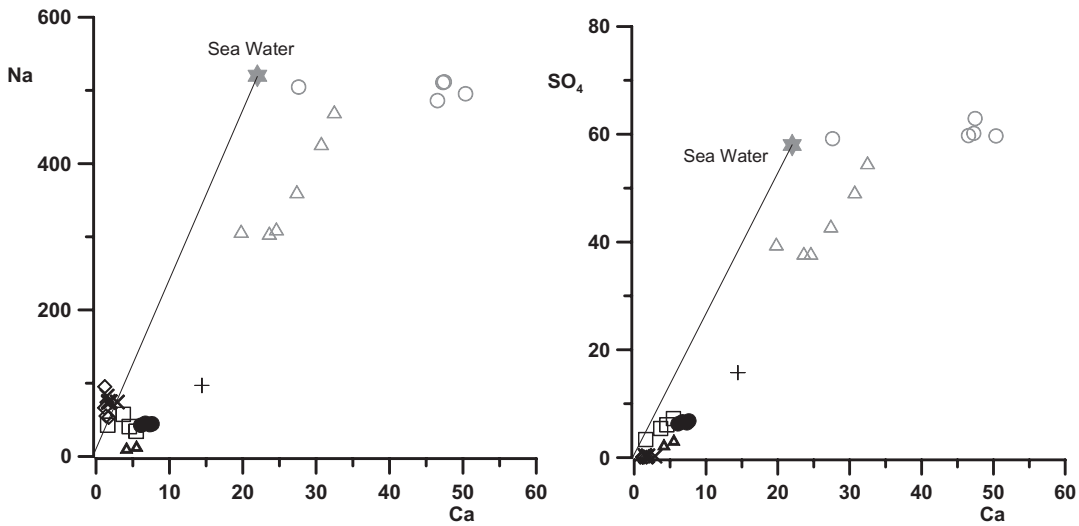


Figure 3 - The Na-Ca (a) and the SO₄-Ca (b) diagrams account for Water-Rock Interactions (WRI) processes. Although the circulating waters are a mixture at various extents of two end members (low salinity component and sea water) the Calcium content is well above that of the sea water showing that a leaching from the local volcanic rocks occurs probably inside thermal aquifers. As the Ca extraction is recognizable in both cold and thermal waters, the possibility that also the cold waters are contaminated by thermal waters was considered.

The Ca-Na and Ca-SO₄ diagrams of figure 3 (a and b respectively) provide further indication on mixing phenomena. As shown by the compositional data of the repeated sampling, both cold and thermal waters underwent modifications of the water chemistry during the sampling period as a probable consequence of changes in the mixing percentage of the two end-members.

Following the geochemical features of the thermal waters, the sea water is here considered as the main fluid recharging a local geothermal aquifer and moreover, their isotope composition (in terms of oxygen and deuterium) are significantly different from the cold waters and from local rain waters. Figure 4 shows the $\delta^{18}\text{O}$ - δD relationships of all the waters collected on the island of Salina besides some data from rain gauges of the nearby island of Stromboli. The graph clearly shows how the thermal waters have different origin from local rainfalls, being the positive values of both oxygen and deuterium isotopic ratios of typical seawater origin.

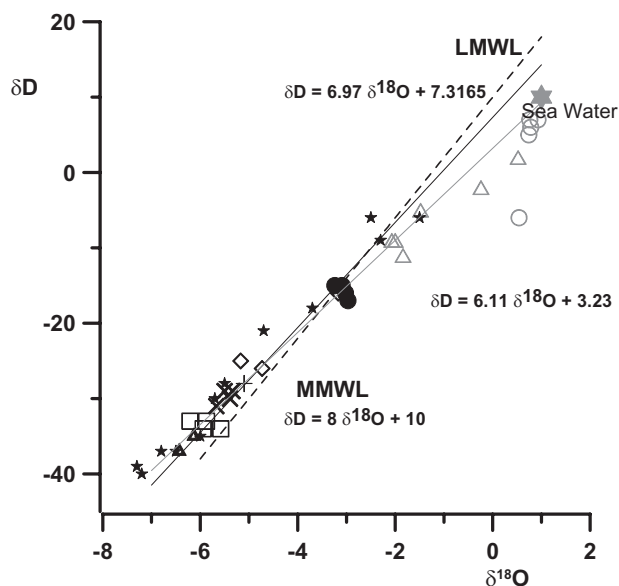


Figure 4 - $\delta^{18}\text{O}$ - δD relationships of the waters from the island of Salina. The black stars are data of rain waters collected at some rain gauges of the nearby island of Stromboli. The regression line of the Salina waters (grey line) and the curves of the dashed and the solid black lines are the local meteoric water line (LMWL) and the Mean Mediterranean meteoric water line (MMWL). The graph shows how the thermal waters have different origin from local rainfalls, being the positive values of both oxygen and deuterium isotopic ratios of seawater origin. A thermal/cold waters mixing is recognizable.

3.2 The dissolved gases

All of the sampled waters have been processed in the laboratory for dissolved gas extraction. The extracted gases have been analyzed for their chemical and isotopic composition with the aim to investigate their origin and to discriminate possible gas-water interaction processes. Starting from the gas-chromatographic analyses, the composition of the dissolved gas phase was calculated taking into account the solubility coefficients (Bunsen coefficient “ β ”, ccgas/mlwater STP) of each gas specie, the volume of extracted gas and that of the water sample, following equation (1):

$$GC = \{[G_{gc}] * V_{\gamma e} + ([G_{gc}] * \beta_G * VW)\} VW^{-1} * V_{\gamma e} * V_{\gamma i}^{-1} / 100 \quad (1)$$

where GC is the concentration of the selected gas specie, G_{gc} is its concentration measured by the gas chromatograph (vol%), $V_{\gamma e}$ and $V_{\gamma i}$ represent the extracted and the introduced gas amount respectively, while VW is the volume of the analyzed water sample (see also Italiano et al., 2004 and 2010 for further details).

The chemical composition of the gases extracted from all of the collected samples is marked by the presence of atmospheric-derived gases (N_2 and O_2) along with non-atmospheric gases (CO_2 , CH_4).

The results, if compared with the typical composition of the gas phase dissolved in waters (either groundwater or sea water (e.g. see the composition of an Air Saturated Sea Water, ASSW), show very low oxygen content up to three orders of magnitude lower than ASSW and a significant presence of CO_2 up to three orders of magnitude higher than ASSW with a remarkable difference for the dissolved gases from cold and thermal waters. Nitrogen and CH_4 are always present with higher concentration in the cold waters while He content in the thermal waters is an order of magnitude higher than that measured in the cold waters.

To get an insight of the origin of the gas phase, the chemical composition of the dissolved gases has been coupled with the isotopic signatures of both helium and carbon. The $^3He/^4He$ ratio, expressed as R/Ra values ($Ra = \text{atmospheric } ^3He/^4He \text{ ratio} = 1.39 \times 10^{-6}$) is plotted versus the $^4He/^{20}Ne$ on Figure 5 together with data of submarine gas samples from the other islands of the Aeolian archipelago [9]. The diagram shows that the $^3He/^4He$ ratios measured in the gases dissolved in the thermal waters of Salina, fall on the same range of the bubbling gases collected at the sea bottom off Pollara, considered to represent the local magmatic signature for the volcanic fluids feeding Salina island. A 4He contamination affects all the cold waters showing $^3He/^4He$ values even below the atmosphere ($0.48Ra$). 4He is probably released by radioactive elements enriched in the old, fractionated, magmatic products. It is worth of notice that all the cold waters had been collected over the oldest portion of the island (SE area). The most recent volcanic activity, in fact (13000 years or less; [2]), occurred on the NW portion of the western edifice that is the youngest of the two volcanoes.

As a result the CO_2 -dominated dissolved gas phase is of magmatic origin, probably released by cooling magmas that are also the thermal source of the geothermal reservoirs located beneath the Island.

3.3 Gas-Water Interactions

The analytical results of the both waters and dissolved gases highlight that the geochemical features of the fluids circulating over the island of Salina are consistent with strong interactions between the liquid and the gas phase. The cold and marine waters are usually equilibrated with the atmosphere (ASW and ASSW) however the high CO_2 and the low O_2 contents show the occurrence of gas-water interactions (GWI) between ASW/ASSW and a CO_2 -dominated gas phase of deep origin. Such an interaction deeply modify the chemical and isotopic composition of both the atmospheric-derived and the deep, magmatic, gas assemblages. The samples plotted on the He- N_2 - CO_2 diagram of figure 6, fall on a CO_2 -dominated/Atmospheric-type mixing line where the thermal waters display the highest CO_2 and helium contents. Besides the CO_2 addition process, the enhanced He and N_2 contents highlight that CO_2 dissolution process occur inside the water bodies.

The slight N_2 excess displayed by the cold waters is interpreted as a local addition of organic-derived gases as the N_2 excess (compared to the normal ASW and ASSW content) couples with the, sometimes very large, CH_4 content.

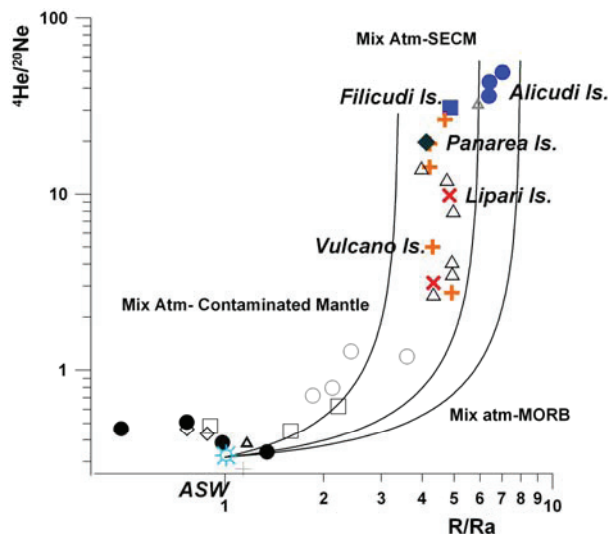


Figure 5 – R/Ra vs He/Ne ratios of helium (normalized to the atmosphere) dissolved in waters from the island of Salina.. The samples show a mixing between the atmosphere and a magmatic-type end member, where magmatic products can be originated either by a contaminated or SECM (Sub European Continental Mantle). The mixing boundary lines are built with the following end members: atmosphere (as dissolved air) R/Ra = 1 and He/Ne = 0.285; Contaminated Mantle R/Ra = 3.4 and He/Ne = 1000; SECM R/Ra = 6.5 and He/Ne = 1000. The graph plots samples from the other Aeolian islands for comparison.

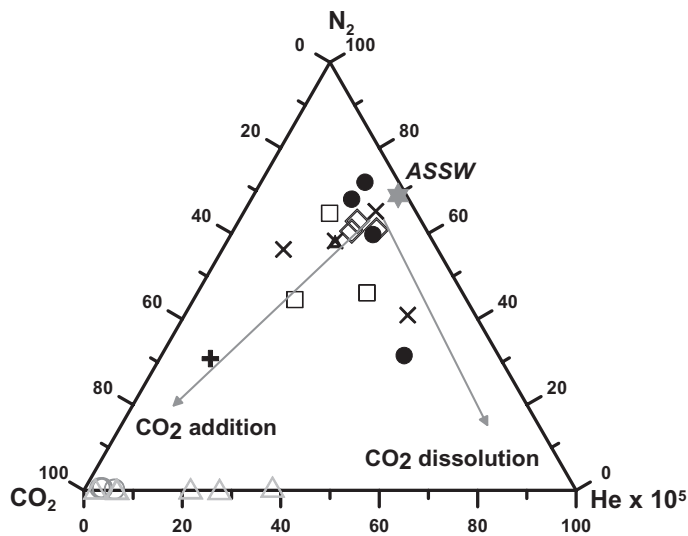


Figure 6 - The triangular He-N₂-CO₂ diagram shows the CO₂ enrichment of all the samples to the respect of the air-saturated sea water (ASSW). Besides the CO₂ addition process, an enhanced He content due to CO₂ dissolution is recognizable. Black marks = cold waters, grey marks = thermal waters; CO₂ content of the thermal waters divided by 10. The slight N₂ excess displayed by the cold waters is interpreted as a local addition of organic-derived gases as the N₂ excess (compared to the normal N₂ content of ASW and ASSW) couples with the CH₄ content (see CH₄ vs CO₂ diagram)

The binary diagrams of figure 7 a, b ($\text{CO}_2\text{-N}_2$ and $\text{CO}_2\text{-CH}_4$) show the occurrence of further processes governing the composition of the dissolved gases. As CH_4 is not a major component of the uprising gases, a local production due to organic matter fermentation is considered as the most probable origin shown by the variable amount of CH_4 in the dissolved gases. Such a process affects only the shallow wells located on the southern coast of the Island where organic-derived carbon is recognizable.

The above mentioned results point out two different effects that contemporary occur: the enrichment of gaseous CO_2 as dissolved phase because of GWI with a CO_2 -rich gas phase, and the CO_2 loss because of new equilibrium inside the water body. As a consequence of GWI, the CO_2 content of the dissolved gas phase increases because of the former process, then its concentration as dissolved gas phase decreases because of the latter. The occurrence of those processes changes both the chemical and isotopic composition of the CO_2 as the heavier carbon (^{13}C) is preferentially enriched in the carbonate ion (CO_3^{2-}).

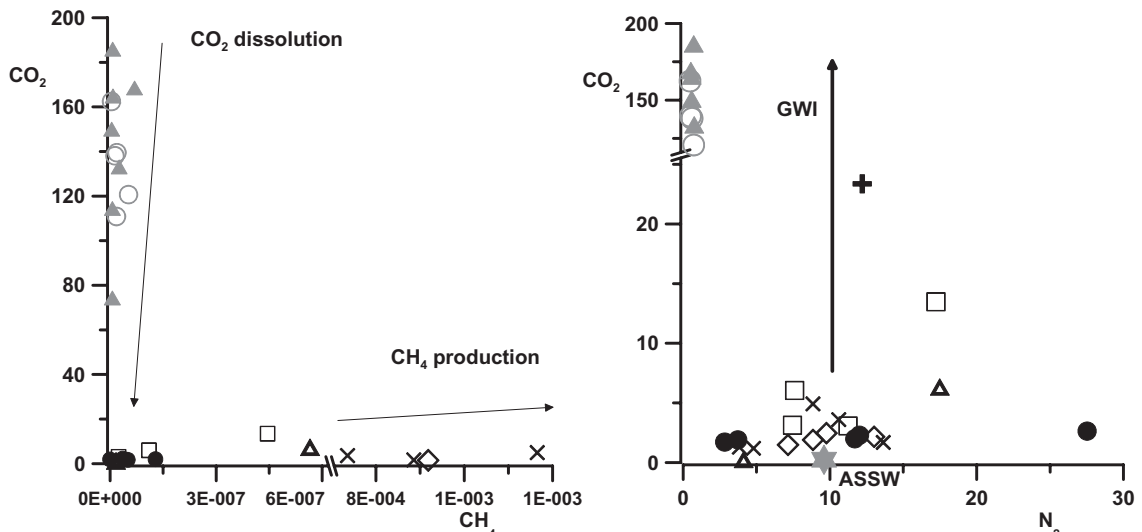


Figure 7 - Gas-Water Interaction processes. GWI (gas-water interaction) processes affecting the composition of gases dissolved in both cold and thermal waters from the Island. The process highlights that an active degassing of magmatic CO_2 affects the island. The released gases interact with the shallow waters (both groundwaters and geothermal bodies) where gases dissolve as a function of their solubility. The circulating waters (marine and fresh) normally carry a dissolved gas phase at the equilibrium with the atmosphere (Air-saturated waters, ASW and air-saturated sea waters, ASSW). The released CO_2 -dominated gases interact with the water bodies allowing CO_2 to dissolve. The thermal waters from the geothermal aquifers are CO_2 saturated, while the shallow cold waters exhibit variable CO_2 content as a function of the GWI intensity 7b - A further process is shown by the variable amount of CH_4 in the dissolved gases. As CH_4 is not a major component of the uprising gases, a local production due to organic matter fermentation is considered as the most probable origin. Such a process affects only the shallow wells located on the southern coast of the Island where organic-derived carbon is recognizable

The isotopic composition of carbon from all the dissolved carbon species (TDC) shows lower values for the cold waters, in the range of $-16.5 < \delta^{13}\text{C} < -6.5\text{‰}$ vs PDB, to the respect of the thermal waters which isotope composition ranges between -2.2 and 0.5‰ vs PDB.

Figure 8 (a, b) plots the $\delta^{13}\text{C}$ values versus CO_2 (fig. 8a) and outlet temperature (fig. 8b). It is evident how the higher is the CO_2 content the higher is the isotopic ratio of carbon, in agreement with GWI processes that enrich the gases in light carbon isotopes (e.g. [10]) and, for the lowest values, with the presence of an organic source contemporary feeding some of the cold waters. The organic carbon is supposed to be of local origin and its presence fits with the large amount of CH_4 dissolved in the cold

waters. The other carbon component (magmatic-type carbon) is degassed across the whole island and dissolves at various depths in the water bodies.

The $\delta^{13}\text{C}$ - T°C diagram shows that the thermal waters bring a dissolved inorganic carbon of deep (magmatic-type) origin as also suggested by the high helium isotopic ratios (up to 5Ra). Contrastingly the low-temperature waters display low carbon isotopic ratios showing the occurrence of enhanced GWI and, sometimes, a mixture of organic and inorganic carbon.

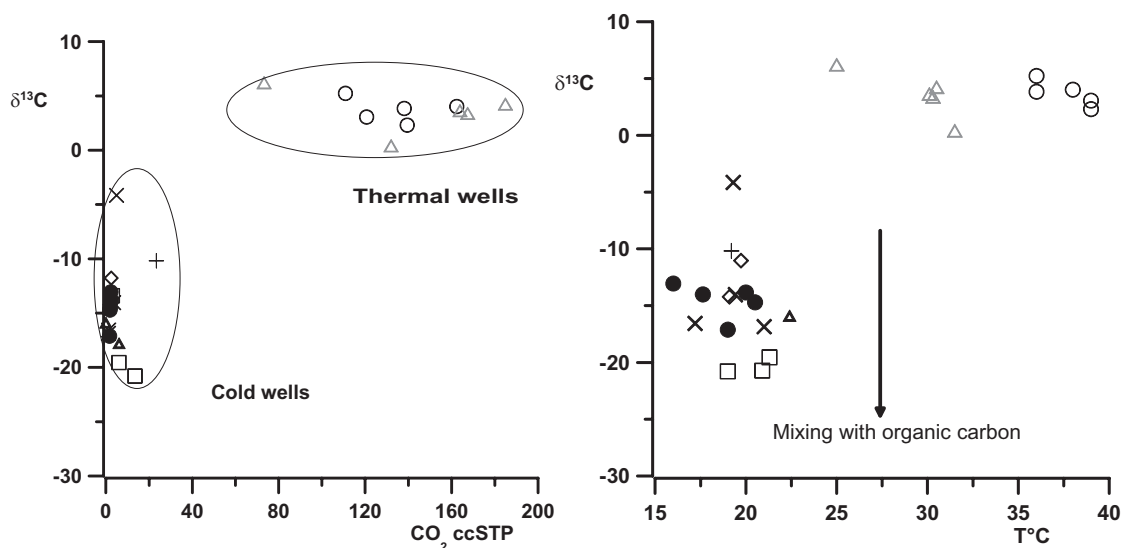


Figure 8 - The $\delta^{13}\text{C}$ - T°C diagram shows that the thermal waters bring a dissolved inorganic carbon of deep (magmatic-type) origin as also suggested by the high helium isotopic ratios (up to 5Ra). Contrastingly the low-temperature waters display low carbon isotopic ratios (even lower than -20‰ vs PDB) showing that those waters bring a mixture of organic and inorganic carbon.

4. Summary and conclusions

The presence of a significant amount of thermal waters, revealed thanks to the drilling of two deep wells, highlights that thermal energy is still available and released by the volcanic rocks of the island. Following the models proposed for the latest evolution of the eruptive activity on the island [1, 2] a basaltic magma chamber existed beneath the island in very recent geologic time. Nowadays, some cooling magma is reasonably available beneath the NW portion of the island, able to thermally feed a relatively deep geothermal aquifer.

Although the circulating waters are a mixture at various extents of two end members (low salinity component and sea water) the Ca content is well above that of the sea water showing that a leaching from the local volcanic rocks occurs probably inside the thermal aquifers. The Na-Ca and the SO_4 -Ca diagrams of figure 3 showing the Ca excess, account for the occurrence of high-temperature Water-Rock Interactions (WRI) processes thus giving strength to the hypothesis that a geothermal aquifer exists. The presence of seawater that suffered high-temperature WRI processes is also in agreement with the increase of the $\delta^{18}\text{O}$ values. The shallow waters (both cold and warm) interact with deep magmatic gases that dissolve in the thermal waters at variable extents as a function of the ratio degassing rate/water body

volume and the length of time of the GWI. All the sampled waters, despite their origin and outlet temperature, bring a dissolved CO₂-dominated gas phase and helium with a clear magmatic signature. As a matter of fact, the investigations carried out on the fluids circulating over the extinct volcano of Salina island show the unexpected presence of two hidden resources: fresh water and geothermal reservoirs. The geochemical features of the fluids revealed that water-rock and gas-water interaction processes occur at various depths changing the original chemical and isotopic composition of the main components: deep magmatic fluids, marine and rain waters.

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