

# Fluid Geochemistry of Stromboli

Fausto Grassa, Salvatore Inguaggiato, and Marcello Liotta

*Istituto Nazionale di Geofisica e Vulcanologia, Sezione di Palermo, Palermo, Italy*

An accurate description of the geochemical system is presented here based on a review of scientific work performed during the past decade. The surface manifestations of the volcanic system of Stromboli have been investigated using several measuring techniques. Studying the chemical composition of the volcanic plume and of fumarolic emissions has provided information on magma degassing processes. The total fluxes of the emitted gases from both the plume and the soil were found to vary with changes in volcanic activity (from normal Strombolian activity to effusive and/or paroxysmal activity). Thermal water results from the interaction between volcanic gases, host rock, seawater, and meteoric water and temporal changes observed in the chemical and the isotopic composition of the gases dissolved into thermal waters highlighted the rising of new magma batches. Combining modeling of gas–water–rock interactions with an understanding of the volcanic system allowed identifying preferential sampling sites and parameters for the geochemical monitoring of volcanic activity at Stromboli Island.

## 1. INTRODUCTION

Geochemical monitoring of active volcanoes provides a useful tool for hazard mitigation. Evaluation of geochemical signals requires a good knowledge of volcanic systems and the formulation of accurate models.

The features of a volcanic system can be defined by its structural and morphological settings, style of volcanic activity, and by the type and the amount of magmatic and/or shallow fluids (e.g., seawater, meteoric water). This geochemical approach can be used to model the interaction processes between the involved fluid phases.

Analyses of volcanic gas, being the most mobile phase in magma, yield useful information on the feeding system. Each volcano presents different degassing styles depending on its origin and evolution. In open-conduit volcanoes, such as Stromboli, most gas is released from the summit vent. Never-

theless, fumarolic fields and diffuse soil degassing represent other important modes of gas release [*Giammanco et al.*, 1998; *Carapezza and Federico*, 2000; *Varley and Armienta*, 2001; *Aiuppa et al.*, 2004; *Brusca et al.*, 2004]. Gases escaping from the magma along high-permeability pathways interact with groundwater and with the host rocks. Dissolution of acidic gases in water quickly lowers the pH which facilitates water–rock interactions. In areas with high gas fluxes, groundwater rapidly becomes saturated with gas and so any further addition of volatiles results in the deep gas-phase flow through the aquifer without significant interactions. In such volcanic systems, gas–water–rock interactions govern the chemical and isotopic compositions of the circulating fluids.

Rising steam and gas that has separated from magma at depth, may interact with shallow aquifers. Released fluids may condense and/or dissolve into the liquid phase, manifesting as hydrothermal systems and/or mineral waters.

The contribution of magmatic fluids to the hydrothermal systems can be hidden by mixing processes with other non-magmatic fluid components (e.g., meteoric waters, seawater, air, crustal and biogenic gas) or by the chemical and isotopic fractionation occurring during water–gas interactions. Nonetheless, many studies have demonstrated that thermal

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Geophysical Monograph Series XXX

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waters associated with the most important active volcanic systems such as Etna, Vulcano, Vesuvius, Ischia Island, Popocatepetl, and El Chichon, tend to maintain the geochemical information of the deep magmatic source [Allard *et al.*, 1997; Capasso *et al.*, 2001; Federico *et al.*, 2004; Inguaggiato *et al.*, 2000, 2005; Taran *et al.*, 1998]. Therefore, thermal discharges represent interesting sites for surface investigations that may provide significant, additional, and sometimes complementary information about deep degassing dynamics.

The occurrence of thermal waters depends on the aquifer geometry, the geological and stratigraphic setting, the hydraulic properties such as transmissivity and porosity, and the recharge and discharge mechanisms. Therefore, the knowledge of the hydrogeological setting is critical to understanding interactions between deep and shallow fluids.

This paper is a review of the most relevant published papers on the fluid geochemistry at Stromboli Island, focusing its main attention on the geochemistry of thermal waters. Based on the data collected since 1999 within the framework of the geochemical volcano monitoring, this paper provides an overview of the most relevant results including a conceptual model of fluid circulation for the volcanic system of Stromboli.

## 2. VOLATILE BUDGET AT STROMBOLI ISLAND

Magmatic volatiles are released during magma migration toward the surface. Decreasing pressure induced by magma batches rising within the conduit and/or by fracturation of the volcanic edifice results in exsolving of fluids whose chemical composition is strongly dependent on the relative solubility of each volatile component in the magma. Generally, volatiles exsolved from magma can be released via the following three main degassing processes:

1. *Open-conduit degassing.* Fluids rising along the conduit reach the top of the magmatic column to form the so-called volcanic plume;

2. *Fumaroles and diffused soil degassing.* Gas and steam are dispersed through the soil as they migrate along high permeability zones;

3. *Degassing associated with geothermal waters.* During their rising, steam and gas may condense and/or dissolve in shallow fluids such as groundwater giving origin to hydrothermal aquifers.

In terms of mass balance, the relative contribution of each degassing process is mainly dependent on the type of volcanism and the level of volcanic activity.

### 2.1. Open-Conduit Degassing

Stromboli is an open-conduit basaltic volcano characterized by persistent degassing and frequent mild explosions

(Strombolian activity). Both quiescent degassing and explosive events contribute to feed a volcanic plume which in the past was discontinuously measured. It has been estimated that the largest fraction of the gas output occurs from the plume, whose total output of gas has been estimated approximately at  $3 \times 10^6$  Mg year<sup>-1</sup> [Allard *et al.*, 1994, this volume]. During persistent passive degassing, the plume chemistry is mainly made of a water-rich gas phase with CO<sub>2</sub>/SO<sub>2</sub> and SO<sub>2</sub>/HCl molar ratios close to 8 and 0.7, respectively [Allard *et al.*, 1994].

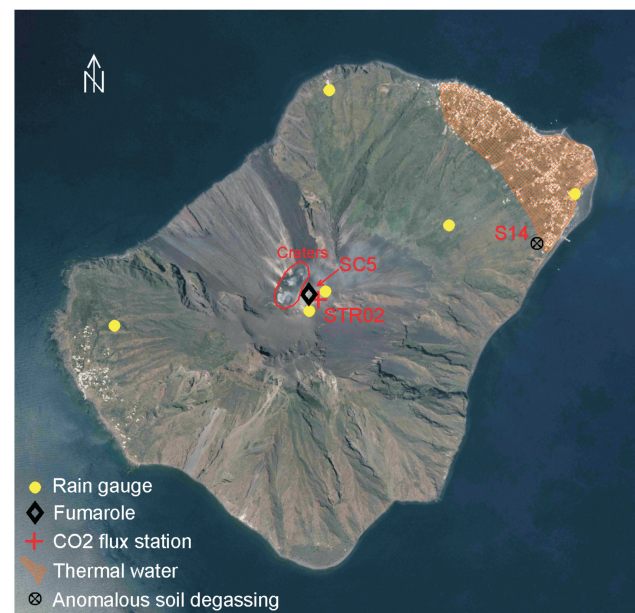
Comparison between CO<sub>2</sub> efflux from the craters and soil in the crater rim area [Carapezza and Federico, 2000] revealed that more than 90% of the gas is released from the open-conduit degassing system.

The mass output of specific gases in crater plume emissions as well as the plume chemistry are continuously measured within the framework of geochemical monitoring of the volcanic activity since this information has provided insights into the plumbing system. For example, the daily SO<sub>2</sub> flux at Stromboli is highly variable with strong increases being evident during 2002–2003 eruption as well as during intense phases of Strombolian activity [Allard *et al.*, this volume]. The plume chemistry is monitored by remote measurements of halogens, sulfur, and carbon compounds [Oppenheimer, 2003]. Within the framework of the geochemical monitoring of volcanic activity at Stromboli 2 d before the 5 April 2003 paroxysm, Aiuppa and Federico [2004], using diffusive tubes, recorded a peak in the SO<sub>2</sub>/HCl ratio four to eight times higher than those observed during passive degassing (SO<sub>2</sub>/HCl ratio ~1, Allard *et al.* [1994]). These authors interpreted this change in the plume chemistry as an evidence of a S-rich magma rising within the volcanic conduit toward the surface which was probably later involved in the paroxysm.

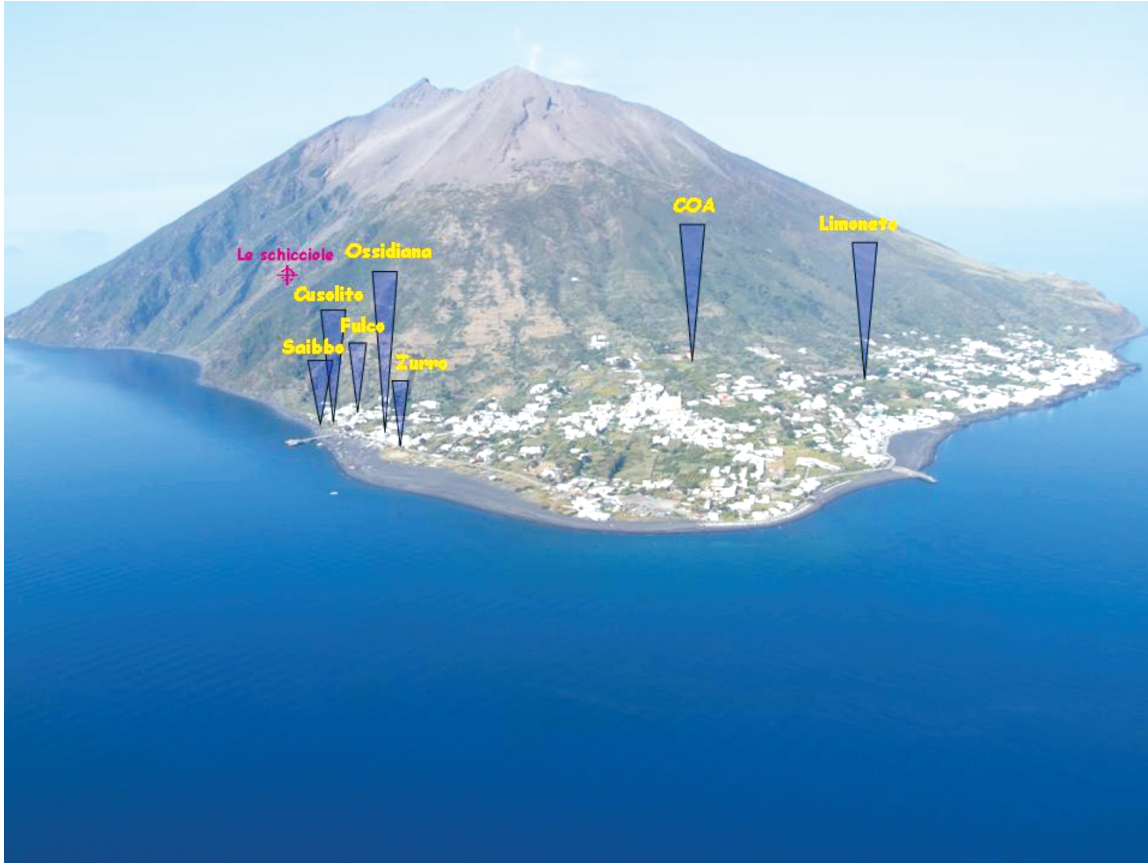
Therefore, the changes in plume composition are useful indicators of the stage of the degassing process and could be used as potential geochemical precursors of high energetic explosions [Aiuppa and Federico, 2004; Rizzo *et al.*, this volume; Allard *et al.*, this volume].

### 2.2. Fumaroles and Diffused Soil Degassing

Uprising hot fluids migrating through highly permeable zones such as porous layers and volcano–tectonic structural discontinuities give rise to hydrothermal activity at the summit area. Finizola *et al.* [2002, 2003, this volume] correlated in situ self-potential measurements with temperature and CO<sub>2</sub> anomalies in several profiles across the entire island. From their results, they proposed a new structural model that includes three perched hydrothermal zones below the summit. These systems, whose structural limits approximately coincide with the local volcano–tectonic structures



**Plate 1.** Base map and sampling sites of natural fluids on Stromboli Island. Filled yellow circles indicate the site of rain gauges; Crater fumaroles (site SC5) as well as low-elevation anomalous soil degassing zones (site S14) are also showed. Site STR 02 is the location of the automatic station installed in July 1999 for continuous monitoring of CO<sub>2</sub> flux from soil (see text). Red grid indicates the northeastern corner of the island where thermal waters were found.



**Plate 2.** Aerial view of Stromboli (taken from northeast) showing the location of thermal wells and Le Schicciolo cold spring.



such as dikes, calderas and regional faults [Finizola *et al.*, 2002, this volume], appear to feed both crateric fumaroles and widespread anomalous diffuse degassing zones at the summit area. Fumaroles are clustered into two zones: (1) in the Fossa area around the eastern flank of the active craters and (2) in the Pizzo area. High-temperature fumaroles (up to 410°C) located in the Fossa area [Martini *et al.*, 1991; Finizola and Sortino, 2003] are sampled only occasionally because this area is highly hazardous due to its high instability and its vicinity to the active vents. In contrast, fumaroles in the Pizzo area are more easily accessible and safe, but they exhibit only low-flux and low-temperature emissions. Carapezza and Federico [2000], Finizola *et al.* [2003], and Capasso *et al.* [2005] found that the maximum temperature was 95°C which is approximately the boiling temperature of water at an elevation of 900 m above sea level (asl).

The data from the most representative fumarole (site SC5, see Plate 1) are listed in Table 1. Despite their low temperature, the chemical and isotope composition of the Pizzo area fumaroles revealed a clear magmatic origin of the gases. CO<sub>2</sub> is the dominant gas phase (up to 93%) having  $\delta^{13}\text{C}_{\text{CO}_2}$  values ranging between  $-3.2$  and  $-0.7$  ‰ versus Vienna Pee Dee belemnite (VPDB). Helium, up to 20 ppmvol, shows a marked mantle-derived signature with the <sup>3</sup>He/<sup>4</sup>He ratios (corrected for air contamination) being in the range between 2.3 and 3.6 R/Ra, where Ra is the <sup>3</sup>He/<sup>4</sup>He ratio in the atmosphere ( $1.39 \times 10^{-6}$ ). The carbon and helium isotope compositions of fumarolic gases at SC5 revealed their precursory character of the 2002–2003 eruption [Capasso *et al.*, 2005; Rizzo *et al.*, this volume; Federico *et al.*, this volume]. A unique field survey at the summit allowed the total output of diffuse CO<sub>2</sub> emissions to be estimated in the range between 200 and 250 t d<sup>-1</sup> [Carapezza and Federico, 2000].

The CO<sub>2</sub> soil efflux in the Pizzo area was monitored by an automatic station (site STR02 see Plate 1) that was installed in July 1999 at a high flux site was considered rep-

resentative of the entire area. The average measured output at this site was a few thousands of grams per square meters per day. Soil gas emissions increased concomitantly with intense explosive activity as well as during preeruptive periods. For instance, the highest soil CO<sub>2</sub> effluxes of about 10<sup>5</sup> g m<sup>-2</sup> d<sup>-1</sup> few were reached a few days before the 2002–2003 eruption [Carapezza *et al.*, 2004; Federico *et al.*, this volume].

Apart from the summit area, CO<sub>2</sub> soil efflux anomalies were also recognized mainly along the northern flank of the edifice where fractures and faults as well as porous layers allow the rising of deep fluids [Finizola *et al.*, 2002] both at intermediate altitudes (Nel Cannestrà and Rina Grande areas) and in the peripheral areas at lower elevations (Pizzillo and San Bartolo). Such anomalous CO<sub>2</sub> soil emissions could be directly related to a deep degassing source (through structural discontinuities), or they may represent a fluid phase separated from thermal reservoirs. Finizola *et al.* [2002] suggested that such anomalous gas emissions are decoupled from the volcanic structures but are linked to two major faults: (1) the N41° structural trend (Pizzillo-Rina Grande alignment) that coincides closely both with a recent dyke intrusion [Bonaccorso, 1998] and a regional fault [Falsaperla *et al.*, 1999] and (2) N64° structural trend (Nel Cannestrà-San Bartolo alignment) that corresponds to a well-known direction of structural weakness zone [Zanchi and Francalanci, 1989].

### 2.3. Thermal Water at Stromboli

Water-well drilling that began in the 1980s first revealed a very shallow hydrothermal system (at a depth of 5–10 m) at nearby Scari village. In this area, there are favorable conditions for the development of an aquifer. The presence of permeable structural elements and porous layers that characterize the entire northern flank [Finizola *et al.*, 2002, this volume] enhances rainwater infiltration. In contrast, the other flanks of the volcano are mainly comprised of scarcely

**Table 1.** Chemical and Isotope Compositions of Low-Temperature (<95°C) Fumaroles at the Summit (Mean, Max, and Min Values)

Sample		He	H <sub>2</sub>	O <sub>2</sub>	N <sub>2</sub>	CO	CH <sub>4</sub>	CO <sub>2</sub>	$\delta^{13}\text{C}_{\text{CO}_2}$	<sup>3</sup> He/ <sup>4</sup> He	He/Ne	<sup>3</sup> He/ <sup>4</sup> He <sub>c</sub>
SC5	Mean	8.9	178.0	4.0	19.6	4.8	15.4	76.5	-2.1	2.3	1.40	2.9
	SD	3.5	173.5	3.6	13.5	7.8	40.7	17.5	0.5	0.4	0.69	0.3
	Max	20.3	770.1	19.0	76.2	45.9	189.3	93.5	-0.7	3.1	3.46	3.6
	Min	4.4	6.2	0.3	6.5	1.1	0.9	2.1	-3.2	1.1	0.45	2.3
	<i>n</i>	38	50	51	51	42	40	51	43	70	70	64
Air		5.2	n.d.	20.8	78	n.d.	n.d.	0.0	-8	1	0.32	1

SD, standard deviation; *n*, number of samples; n.d., no data. He, H<sub>2</sub>, CO, and CH<sub>4</sub> contents are expressed in ppmVol, while O<sub>2</sub>, N<sub>2</sub>, and CO<sub>2</sub> in volumetric percentages. Helium isotope ratios (<sup>3</sup>He/<sup>4</sup>He) are expressed as R/Ra. <sup>3</sup>He/<sup>4</sup>He<sub>c</sub> is the helium isotope ratio corrected for air contamination [Sano *et al.*, 1993]. Air composition is also given for reference.

**Table 2.** Physicochemical Parameters, Chemical and Isotope Composition of Thermal Waters Collected at Stromboli

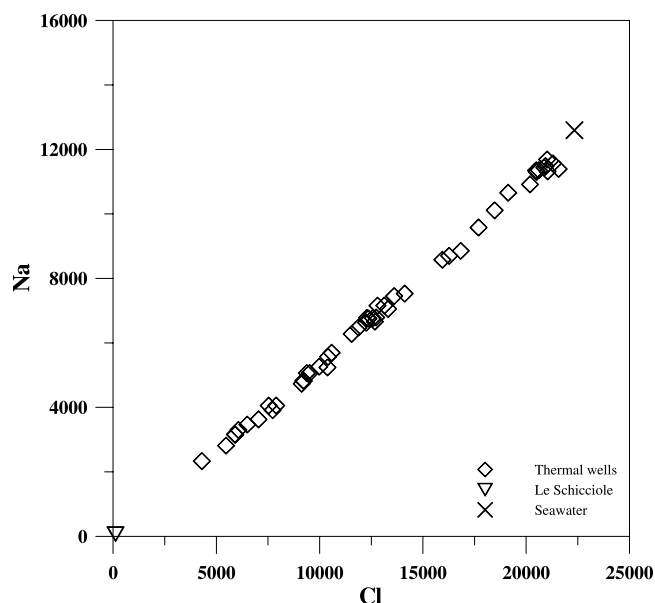
Sample		T, °C	pH	EC, mS		Eh, mv	Na	K	Mg	Ca	F	Cl	Br	NO <sub>3</sub>	SO <sub>4</sub>	HCO <sub>3</sub>	δ <sup>18</sup> O	δD
				cm <sup>-1</sup>														
Zurro	Mean	35.4	6.97	43.3	-38.4	10,704	513	1,200	542	46	19,300	66	72	2,475	315	0.1	1	
	SD	1.1	0.21	7.3	85.6	1,250	87	201	116	145	2,329	14	53	298	41	0.9	4	
	Max	37.5	7.78	55.0	272.3	12,104	842	1,467	856	661	22,351	120	203	2,926	470	0.9	6	
	Min	32.3	6.61	23.3	-236.0	6,030	292	145	181	1	9,974	22	3	1,394	229	-2.0	-9	
	n	125	127	88	98	94	94	94	94	20	94	93	32	94	112	43	32	
Fulco	Mean	41.1	6.41	18.0	21.0	3,373	263	465	302	5	6,053	20	32	938	791	-4.2	-22	
	SD	1.2	0.11	8.4	85.4	1,844	85	194	92	5	3,450	11	22	376	133	1.1	6	
	Max	43.1	6.98	39.9	242.0	7,980	484	966	572	18	14,881	71	103	1,929	1,104	-2.0	-10	
	Min	37.6	6.19	-13.7	-266.0	782	123	190	121	1	11	3	5	360	458	-5.8	-34	
	n	116	113	110	103	101	101	101	101	100	101	101	99	101	109	53	42	
Saibbo	Mean	39.9	6.31	30.5	86.3	7,134	428	880	523	n.d.	13,267	44	n.d.	1,717	474	-2.0	-11	
	SD	2.5	0.11	5.6	84.7	1,122	51	132	65	-	2,132	7	-	293	47	0.6	3	
	Max	42.6	6.55	37.8	237.0	9,579	555	1,125	668	-	17,698	62	-	2,315	528	-0.8	-5	
	Min	29.9	6.02	7.9	-80.0	5,596	377	683	425	-	10,539	35	-	1,311	305	-2.9	-15	
	n	26	26	26	21	17	17	17	17	-	17	17	-	17	25	12	12	
Limoneto	Mean	41.0	6.66	29.9	11.7	6,537	459	871	439	n.d.	12,263	39	n.d.	1,591	650	-2.9	-16	
	SD	1.1	0.07	6.4	105.7	1,538	61	154	70	-	2,766	10	-	351	114	1.0	5	
	Max	42.8	6.8	38.8	283.5	9,509	582	1,206	603	-	17,902	58	-	2,295	885	-0.7	-10	
	Min	38.6	6.53	17.6	-129.0	3,626	343	593	313	-	7,040	19	-	963	464	-4.8	-24	
	n	32	32	29	26	37	37	37	37	-	37	37	-	37	42	23	19	
COA	Mean	40.5	6.52	27.6	30.5	5,944	369	770	431	n.d.	11,011	36	n.d.	1,393	448	-2.2	-12	
	SD	2.1	0.08	4.9	70.1	1,821	125	224	79	-	3,254	10	-	412	151	-	-	
	Max	42.1	6.62	31.5	78.0	7,231	458	929	487	-	13,313	43	-	1,684	555	-	-	
	Min	38.1	6.47	22.1	-50.0	4,656	281	612	376	-	8,710	29	-	1,101	342	-	-	
	n	3	3	3	3	2	2	2	2	-	2	2	-	2	2	1	1	
Cusolito	Mean	42.3	6.68	33.7	-77.1	7,682	448	883	484	11	14,023	47	56	1,792	494	-2.0	-12	
	SD	1.4	0.18	5.8	45.9	1,745	90	208	114	11	3,326	13	53	374	73	0.7	4	
	Max	46.9	7.18	45.4	32.0	10,776	605	1,209	724	42	19,969	76	228	2,410	708	-0.6	-8	
	Min	38.9	6.27	17.0	-205.8	3,198	284	473	264	1	5,994	14	1	770	305	-3.6	-20	
	n	62	62	61	57	65	65	65	65	11	65	65	26	65	65	23	11	
Ossidiana	Mean	35.8	6.97	36.0	-88.9	9,106	416	943	457	2	16,026	54	26	2,068	604	n.d.	n.d.	
	SD	1.8	0.14	20.7	97.3	4,491	131	385	113	1	7,685	26	5	965	196	-	-	
	Max	38.3	7.7	61.5	169.9	12,822	559	1,270	617	4	22,013	83	32	3,041	772	-	-	
	Min	31.9	6.78	9.8	-264.0	1,858	209	325	276	1	3,572	9	20	611	302	-	-	
	n	73	70	41	34	20	20	20	20	20	20	20	4	20	46	-	-	
Le Schicchiole	17/04/03	15.9	6.62	1.1	n.m.	63	30	30	159	7	119	n.d.	n.d.	403	403	-6.2	-34	
Seawater	23/05/07	21.1	8.14	42.9	98.0	11,648	415	1,477	487	n.d.	21,798	76	n.d.	171	171	0.6	6	

All data are reported in terms of Min, Max, and Min values. SD, standard deviation; *n*, number of samples; n.d., data. Data relative to the Le Schicchiole cold spring sample and a seawater sample collected in proximity of the coast of Stromboli Island are also given for comparison. Ion concentrations are expressed in mg kg<sup>-1</sup>. Isotope values are expressed in parts per thousand (‰) with respect to the Vienna standard mean ocean water (VSMOW) international reference.

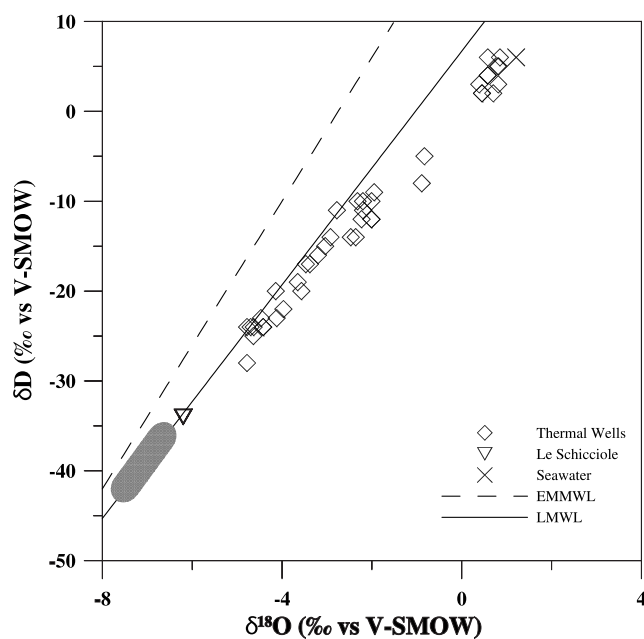
permeable deposits or sealed altered lavas that result in low infiltration rates. For these reasons, exploration drilling aimed at finding thermal waters in the remnant part of the island was unproductive.

Nowadays, there are about 10 thermal wells clustered in a limited area on the northeastern corner of Stromboli (Plate 2), two of which were drilled for scientific purposes in 2004

(Saibbo well) and 2005 (COA well). Thermal wells are located at elevations between 5 and 70 m asl. Their respective phreatic levels are reasonably constant and slightly lower than the seawater level. This suggests the occurrence of unconfined or partially confined aquifers hosted within a spatially homogeneous porous media (i.e., pyroclastic deposits) that are hydraulically isolated from seawaters probably due



**Figure 1.** Binary diagram showing Na versus Cl contents in the groundwater of Stromboli. All the samples fall along the seawater ratio line. Concentrations are expressed in  $\text{mg kg}^{-1}$ . Selected data from the INGV-PA internal database.

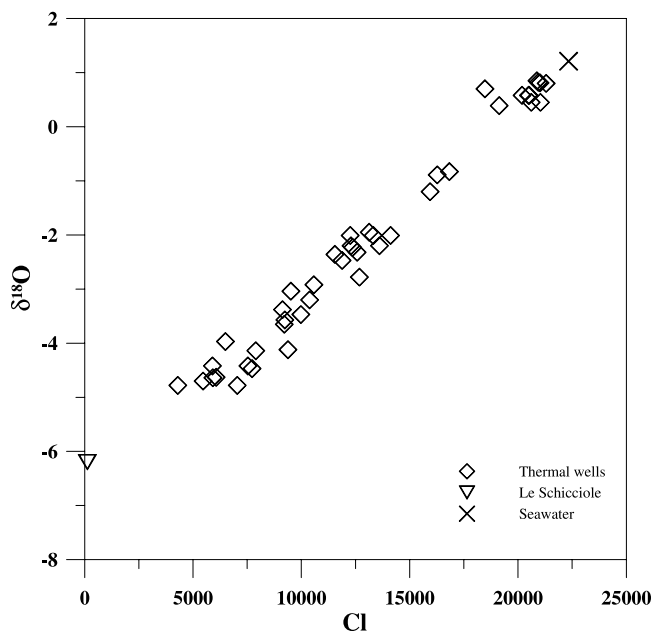


**Figure 2.**  $\text{Cl-SO}_4\text{-HCO}_3$  ternary diagram. All the collected waters show a trend from seawater composition toward that of Le Schicciolo sample. In volcanic areas, the  $\text{HCO}_3$ -rich end-member has been related to peripheral waters [Giggenbach, 1991] resulting from the interaction between groundwater and magmatic  $\text{CO}_2$ . Selected data from the INGV-PA internal database.

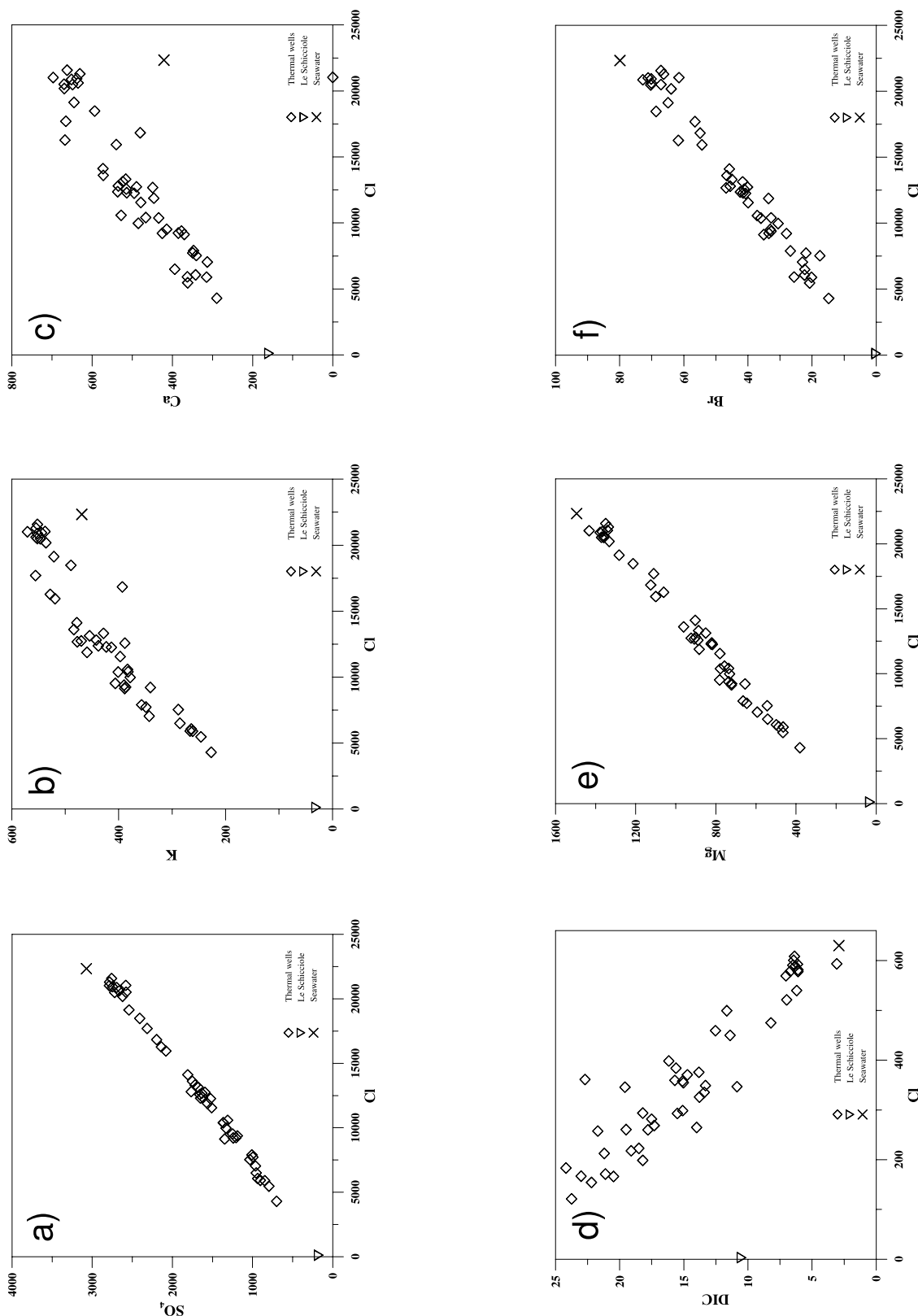
to the presence of impermeable layers such as altered lava flows.

### 3. GEOCHEMICAL INVESTIGATIONS ON THERMAL WATERS

In this section is reported an overview of the most relevant results concerning a pluriannual geochemical monitoring of thermal waters at Stromboli. The thermal waters are periodically sampled to assess the magmatic contribution of fluids and heat to the thermal aquifers and to model the gas–water–rock interactions in the thermal aquifers of Stromboli. Seawater and freshwater from the only one cold discharge at Stromboli island (Le Schicciolo cold spring) were also collected. The concentration of the major and some of the minor dissolved elements and the chemical composition of the main dissolved gas phase were determined in the collected groundwater. Moreover, a rain gauge network consisting of six stations was installed to collect suitable samples for chemical and isotope analyses (Plate 1). Precipitation was sampled approximately bimonthly between October 2003



**Figure 3.** Diagram showing  $\delta\text{D}$  versus  $\delta^{18}\text{O}$  in the groundwater of Stromboli. Open symbols indicate thermal (squares) and shallow (diamonds) groundwater samples. The dotted area encloses the meteoric recharge composition [Liotta *et al.*, 2006]. The solid line is the local meteoric water line (LMWL, Liotta *et al.* [2006]), and the dashed line is the Eastern Mediterranean meteoric water line (EMMWL [Gat and Carmi, 1970]). Selected data from the INGV-PA internal database.



**Figure 4.** Correlation plot of  $\delta^{18}\text{O}$  versus Cl. All samples fall between two end-members: seawater and shallow groundwater. Concentrations are expressed in  $\text{mg kg}^{-1}$ , and isotope values are reported in  $\delta$  values versus Vienna standard mean ocean water (VSMOW).

and October 2005 [Liotta *et al.*, 2006]. Finally, the  $\delta D$  and  $\delta^{18}O$  values of waters,  $\delta^{13}C_{CO_2}$  values and  $^3He/^4He$  ratios in the gas phase dissolved in thermal waters have also been measured [Carapezza and Federico, 2000; Capasso *et al.*, 2005, unpublished data from INGV-PA internal database].

### 3.1. Water Geochemistry

Physico-chemical parameters and the chemical and the isotope composition of waters collected from five selected thermal wells (Fulco, Limoneto, Saibbo, COA, and Zurro) are listed in Table 2. This table also lists analytical data relative to the Le Schiccirole (LS) cold spring sample (temperature = 15.9°C) and are also shown together with a seawater sample collected near the beach of Punta Lena. Data are extracted from the INGV-PA internal database but those from 1999 to 2000 and from 2002 to 2003 which were published by Carapezza and Inguaggiato [2001] and Capasso *et al.* [2005], respectively.

The water temperature which was almost constant over time, ranged from 35.5°C (Zurro) to 42.8°C (Fulco). The pH was nearly neutral or slightly acidic ranging between 6.3 (Saibbo well) and 6.9 (Zurro well). The water chemistry was dominated by NaCl with an almost constant Na/Cl ratio over time and that was close to that of the seawater (Figure 1). The total dissolved solids (TDS) content varied from 7.1 g l<sup>-1</sup> (Fulco) to about 40 g l<sup>-1</sup> (Zurro).

In the Cl–HCO<sub>3</sub>–SO<sub>4</sub> ternary diagram in Figure 2, all the collected waters show a trend from seawater composition toward the composition of the Le Schiccirole sample. The latter sample falls within the “peripheral waters” field typical for waters that reflect the dissolution of CO<sub>2</sub>-rich magmatic fluids into meteoric waters [Giggenbach, 1991]. Due to its relatively low salinity, it is a sample representative of a low degree of gas–water interaction, as better described in the following subsection.

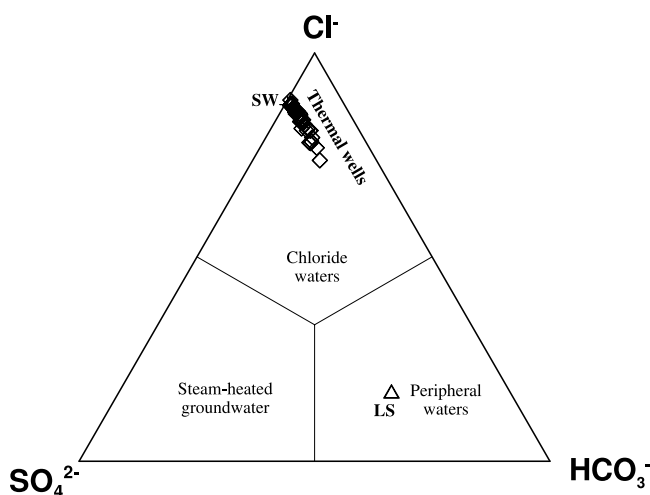
Stable isotope data of collected waters are plotted on a  $\delta D$ – $\delta^{18}O$  diagram (Figure 3), which also includes the Mediterranean meteoric water reference line (MMWL  $\delta D = 8 \times \delta^{18}O + 22$ , Gat and Carmi [1970]) and the local meteoric water line (LMWL,  $\delta D = 6.5 \times \delta^{18}O + 6.7$ , Liotta *et al.* [2006]).  $\delta D$  and  $\delta^{18}O$  values range from –34‰ (Fulco) to +6‰ (Zurro) and from –5.8‰ (Fulco) to +0.9‰ (Zurro), respectively. All the sample points appear on a mixing line between the seawater composition ( $\delta D = 6$ ‰ and  $\delta^{18}O = 0.6$ ‰) and the composition of mean annual weighted precipitation ( $\delta D \approx -40$ ‰ and  $\delta^{18}O \approx -7.2$ ‰) at Stromboli [Liotta *et al.*, 2006], thus suggesting that the thermal waters result from mixing in different proportions between these two end-members. This process is also indicated by the strong correlation between  $\delta^{18}O$  and Cl contents (Figure 4).

### 3.2. Water–Rock Interaction Processes

Figure 5 plots the SO<sub>4</sub>, Ca, K, DIC (i.e., dissolved inorganic carbon), Mg, and Br contents in the sampled waters against the Cl content. Although the thermal waters of Stromboli are characterized by a wide range of Cl concentrations and therefore also of salinity, the compositional ratios vary only slightly among the wells. Therefore, thermal waters seem to result from a simple mixing between meteoric-like waters and the saline waters, where the former is similar to seawater.

By assuming that all dissolved Cl has a marine origin and that it behaves conservatively, meaning that this element is not involved in other geochemical processes, the hydrothermal saline end-member should have a Cl content equal to seawater (i.e., 22,300 mg kg<sup>-1</sup>). Following the approach used by Taran *et al.* [2002], the total chemical composition of the hydrothermal saline end-member was extrapolated to a Cl concentration of 22,300 mg kg<sup>-1</sup>. The obtained chemical composition was characterized by a slightly lower SO<sub>4</sub> concentrations but much higher Ca, K, and DIC contents than those of seawater with Na, Mg, and Br contents being comparable to those of seawater.

This indicates that the pristine seawater composition is chemically modified by leaching high-K calcalkaline to



**Figure 5.** Major and minor ions plotted versus Cl contents in Strombolian groundwater. Mg/Cl (e), DIC/Cl (d), and Br/Cl (f) reflect a seawater composition, whereas K/Cl (b), Ca/Cl (c), and DIC/Cl (d) differ significantly from the seawater ratio being enriched in K, Ca, and DIC. The SO<sub>4</sub>/Cl ratio (a) is slightly lower than that of seawaters. Concentrations are expressed in mg kg<sup>-1</sup>, except for panel (d) which is in mmol l<sup>-1</sup>. Selected data from the INGV-PA internal database.



shoshonitic volcanic rocks of the aquifer and weathering of plagioclase that in Strombolian rocks have 45 to 90% anorthite content [Francaianci, 1993]. On the contrary,  $\text{SO}_4$  may be involved in reduction reactions favored by the prevailing reduction condition in the thermal aquifer.

Similarly, we have also estimated the chemical composition of the diluted end-member. In this case, we extrapolated to a Cl content of 117 mg  $\text{kg}^{-1}$  which is the average Cl content in the rainwater collected at the summit crater [Liotta *et al.*, 2006]. The obtained composition roughly coincides with that of the Le Schicciolate cold spring. The most relevant

differences between the inferred and measured Le Schicciolate compositions are in the depletion both in DIC and in K contents. These differences could be due to a lower degree of interaction between meteoric recharge and  $\text{CO}_2$ -rich deep fluids leading to reduced leaching of rocks as well.

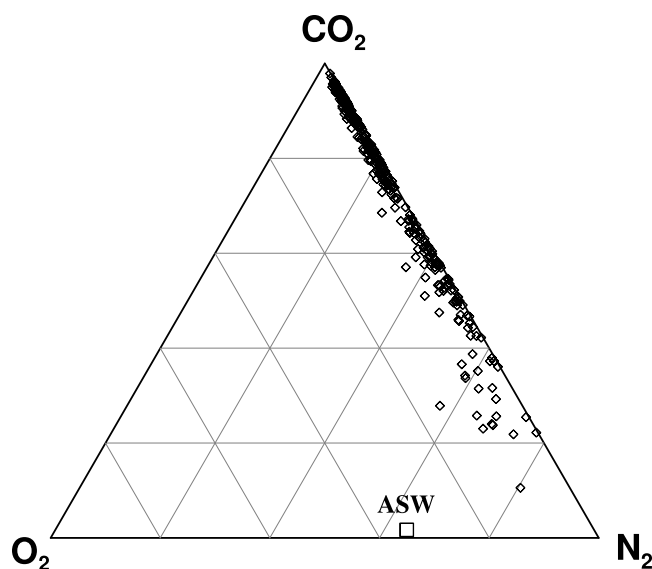
### 3.3. Dissolved Gas Geochemistry

Table 3 reports the concentrations of dissolved gaseous species expressed in cubic centimeters per liter ( $\text{cc l}^{-1}$ ) of water at standard temperature and pressure (STP) condition

**Table 3.** Chemical and Isotope Compositions of Gas Dissolved in Thermal Wells Reported in Terms of Mean, Max, and Min Values

Sample		He	H <sub>2</sub>	O <sub>2</sub>	N <sub>2</sub>	CO	CH <sub>4</sub>	CO <sub>2</sub>	$\delta^{13}\text{C}_{\text{TDC}}$	$^3\text{He}/^4\text{He}$
Zurro	Mean	7.62E-04	1.01E-02	0.6	15.0	2.22E-04	6.44E-03	20.3	-0.8	4.16
	SD	8.34E-04	2.87E-02	0.7	3.4	2.27E-04	9.67E-03	19.3	1.9	0.14
	Max	6.47E-03	2.04E-01	3.2	29.1	1.18E-03	8.34E-02	140.9	4.5	4.41
	Min	1.48E-04	3.39E-04	0.1	3.6	7.94E-06	1.83E-04	1.8	-5.8	3.74
	<i>n</i>	61	64	109	127	68	121	127	108	91
Fulco	Mean	8.58E-04	7.22E-03	0.9	11.5	4.21E-04	4.44E-03	159.6	1.3	4.16
	SD	1.93E-03	1.86E-02	0.6	2.7	4.23E-04	4.22E-03	35.0	0.9	0.16
	Max	1.58E-02	1.02E-01	3.0	19.2	2.28E-03	3.51E-02	292.3	1.6	4.49
	Min	1.73E-04	2.19E-04	0.1	5.6	1.89E-05	9.06E-05	76.1	-0.6	3.46
	<i>n</i>	64	55	122	129	53	112	129	123	88
Saibbo	Mean	7.36E-04	7.14E-03	0.6	12.3	3.19E-04	4.95E-03	129.4	0.3	4.24
	SD	2.39E-04	1.49E-02	2.2	1.5	3.71E-04	1.14E-03	23.7	0.6	0.09
	Max	1.38E-03	5.90E-02	13.8	16.6	1.71E-03	6.46E-03	183.5	2.0	4.44
	Min	2.56E-04	3.05E-04	0.0	8.6	2.54E-05	1.28E-03	62.8	-0.8	4.06
	<i>n</i>	39	25	39	39	37	39	39	32	40
Limoneto	Mean	1.27E-03	8.15E-03	0.6	13.6	1.26E-04	7.67E-03	66.4	0.8	4.22
	SD	5.47E-04	1.24E-02	0.4	2.1	2.00E-04	4.76E-03	17.2	0.7	0.08
	Max	2.90E-03	4.15E-02	1.3	20.2	8.81E-04	2.24E-02	94.5	2.2	4.43
	Min	6.04E-04	4.05E-04	0.0	6.4	8.73E-06	1.93E-03	8.1	-1.0	4.07
	<i>n</i>	39	25	37	38	17	39	39	41	40
COA	Mean	1.13E-03	4.09E-03	0.1	14.6	5.00E-05	1.87E-02	83.5	-0.4	4.24
	SD	1.65E-04	5.10E-03	0.1	1.5	6.80E-05	1.28E-02	12.0	0.3	0.07
	Max	1.35E-03	1.56E-02	0.5	17.0	1.87E-04	4.00E-02	104.4	0.3	4.37
	Min	8.30E-04	3.94E-04	0.0	12.7	7.14E-06	7.01E-03	66.8	-0.8	4.08
	<i>n</i>	11	9	11	11	6	11	11	7	14
Cusolito	Mean	9.95E-04	1.86E-02	0.8	14.7	2.10E-04	4.65E-03	58.3	1.2	4.16
	SD	1.59E-03	5.26E-02	0.6	3.0	1.55E-04	6.33E-03	30.1	1.2	0.16
	Max	8.56E-03	2.83E-01	3.1	25.9	7.12E-04	4.09E-02	219.1	4.9	4.50
	Min	1.99E-04	1.29E-04	0.1	8.0	4.23E-05	2.39E-04	22.9	-0.4	3.73
	<i>n</i>	26	35	73	75	26	74	75	73	49
Ossidiana	Mean	1.15E-03	7.09E-03	1.7	13.6	1.98E-04	2.95E+00	45.6	1.3	n.d.
	SD	6.37E-04	1.45E-02	1.1	4.4	2.78E-04	9.38E+00	26.8	1.1	n.d.
	Max	2.33E-03	6.79E-02	3.6	27.9	1.12E-03	3.43E+01	86.7	2.7	n.d.
	Min	3.06E-04	3.58E-04	0.1	6.5	2.27E-05	1.30E-04	0.5	-0.1	n.d.
	<i>n</i>	10	22	31	33	26	32	33	16	n.d.
ASW		4.81E-05	n.d.	6.4	12	n.d.	n.d.	0.3	0	1

SD, standard deviation; *n*, number of samples; n.d., no data. He, H<sub>2</sub>, CO, and CH<sub>4</sub> contents are expressed in ppmVol, while O<sub>2</sub>, N<sub>2</sub>, and CO<sub>2</sub> in volumetric percentages.  $\delta^{13}\text{C}_{\text{TDC}}$  values are given in parts per thousand (‰) with respect to the Vienna Peedee belemnite (VPDB). Helium isotope ratios ( $^3\text{He}/^4\text{He}$ ) are expressed as R/Ra. Air saturated water (ASW) composition is also given for reference.



**Figure 6.**  $O_2$ - $N_2$ - $CO_2$  ternary diagram. All the samples are characterized by an  $O_2/N_2$  ratio lower than that of air saturated water (ASW) due to oxygen consumption associated with reduction reactions.

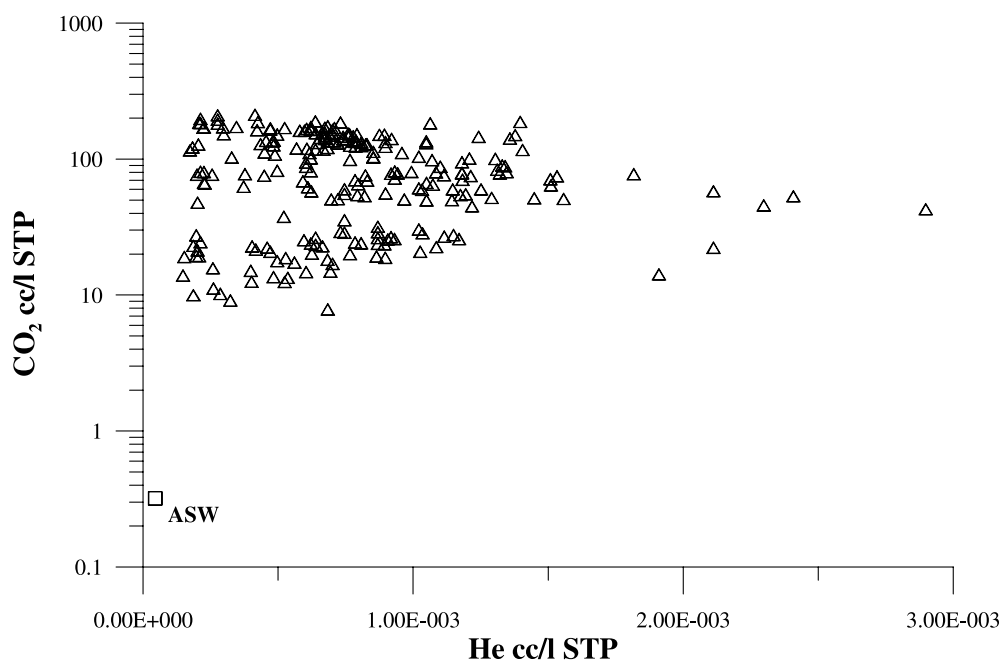
together with the concentration of dissolved gases in a water sample equilibrated with air [air saturated water (ASW)]. This table also lists the carbon isotope composition of the total dissolved inorganic carbon ( $\delta^{13}C_{TDIC}$ ) as well as the helium isotope ratios.

$O_2/N_2$  ratios lower than that of ASW (Figure 6) are probably due to the oxygen consumption associated to reduction processes [Capasso *et al.*, 2005], while high  $CH_4$  contents, as high as  $1.1 \times 10^{-2}$  cc  $l^{-1}$  STP [Capasso *et al.*, 2005] are produced within the hydrothermal systems itself rather than derived from a magmatic source.

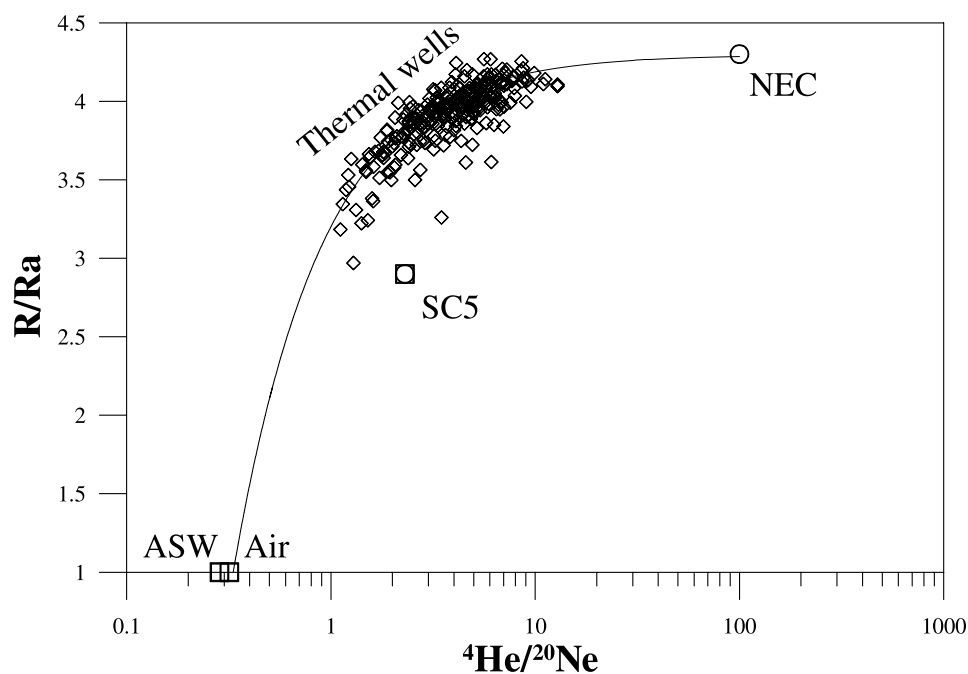
The dissolved helium shows contents exceeding that of ASW by at least one order of magnitude ( $He_{ASW}$  at  $25^\circ C = 4.5 \times 10^{-5}$  cc  $l^{-1}$  STP). The dissolved  $CO_2$  contents are quite high at between 20 and 30 cc  $l^{-1}$  STP (Zurro) and 200 cc  $l^{-1}$  STP (Fulco; Figure 7), mainly due to its high solubility in water ( $\beta_{CO_2}$  at  $25^\circ C = 862$  cc  $l^{-1}$  STP, Whitfield [1978]).

The  $\delta^{13}C_{TDIC}$  values range between  $-5.8$  and  $+4.5\%$  versus PDB. This geochemical parameter represents the average of the isotopic composition of the dissolved inorganic carbon species ( $CO_{2g}$ ,  $HCO_3^-$ , and  $CO_3^{2-}$ ) weighted on their respective content.

When the  $\delta^{13}C_{TDIC}$  values as well as the concentration of each dissolved inorganic carbon species are known, the isotopic composition of the free  $CO_2$  in equilibrium with



**Figure 7.** Correlation plot of He versus  $CO_2$ . Dissolved carbon dioxide and helium contents exceed those of ASW by up to three and up to two orders of magnitude, respectively. Both gases have a clear magmatic origin (see text).



**Figure 8.** Plot of  $R/Ra$  versus  $He/Ne$  molar ratios. All the samples lie on a theoretical mixing curve between an atmospheric component (air or ASW) and the most representative magmatic end-member (NEC sample, *Finizola and Sortino* [2003]). The mean composition at site SC5 is also shown.

thermal waters of Stromboli was computed [*Capasso et al.*, 2005] for an equilibrium temperature of 40°C which corresponds to the mean sampling temperature.

The obtained  $\delta^{13}C_{CO_2g}$  values ranged between  $-4.4\%$  and  $-1.1\%$  versus VPDB. These values are slightly more negative than the carbon isotope composition of the SC5 fumarole, with this difference being attributed to isotope fractionation processes between deep and shallow fluids.

The isotope ratios of helium dissolved in the thermal waters at Stromboli, after correcting for air contamination using the method proposed by *Sano et al.* [1993], range between 3.4 and 4.5 Ra [*Inguaggiato and Rizzo*, 2004; *Capasso et al.*, 2005]. The measured values are very close to those found by *Finizola and Sortino* [2003] in the high-temperature fumarole near the northeast crater formed during the 2003 eruption, whose average value (4.3 Ra) was considered as the magmatic end-member for Stromboli (Figure 8).

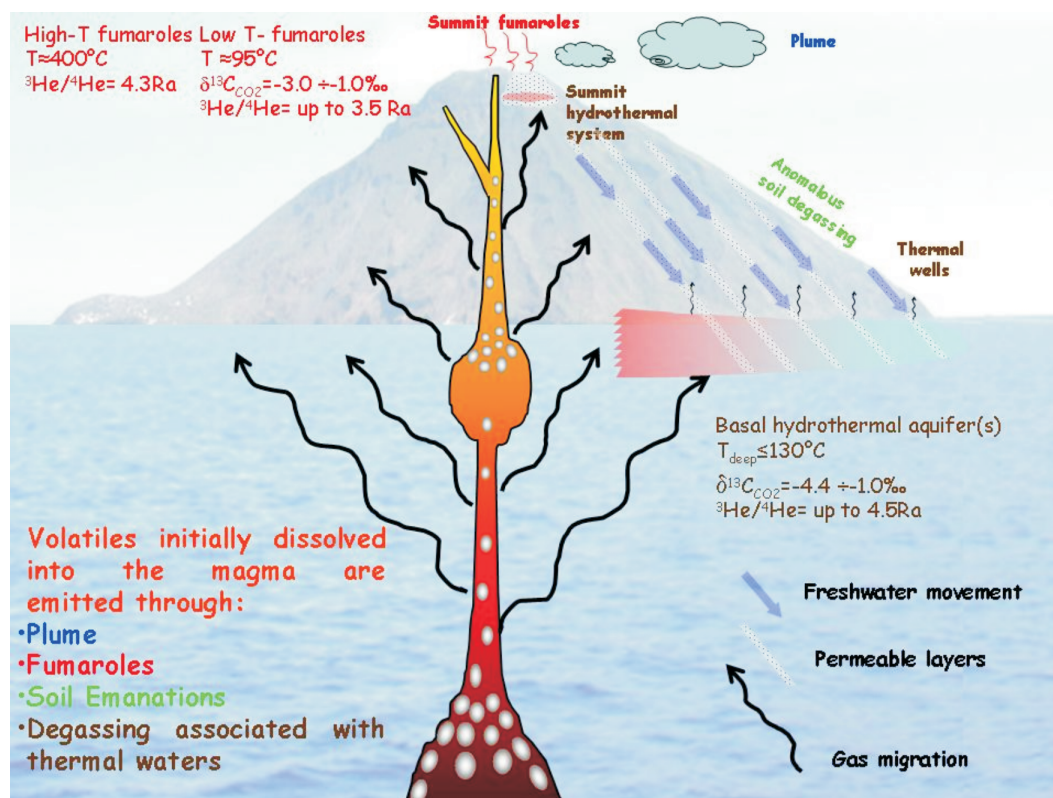
On the basis of the  $He/CO_2$  ratios and the calculated  $\delta^{13}C_{CO_2}$  values in equilibrium with thermal waters, *Capasso et al.* [2005] proposed a geochemical model for the origin of fluids dissolved in the Stromboli thermal aquifer. A vapor phase is partially separated from a boiling aquifer heated by magmatic fluids at a temperature of about 130°C and 2.5 bars. Rising fluids that separate from the boiling aquifer condense in the shallow aquifer at about 40°C.

Therefore, gas phase dissolved in thermal waters of Stromboli derived from magmatic volatiles and helium isotope ratios in the dissolved gas provide the same geochemical information as the high-temperature crater fumaroles.

Temporal variations observed in the helium isotope ratios dissolved in the thermal waters of Stromboli, revealed a clear uprising of new batches of  $^3He$ -rich magma, a few months preceding the 2002–2003 eruption [*Capasso et al.*, 2005].

#### 4. CONCLUDING REMARKS

Chemical and isotopic data on fluids discharged from the Stromboli volcano allow the main geochemical processes affecting the deep fluids during their rise toward the surface to be identified. Moreover, the composition of the end-members feeding the geothermal system has been calculated. Plate 3 shows the formulated geochemical model explaining the fluid circulation. This proposed model considers Stromboli volcano as a natural system where the totality of energy and mass of deep fluids coming from magmatic chamber is discharged at the surface following different pathways. The volatiles initially dissolved into the magma are emitted via the plume, fumaroles, soil emanations, and as fluids associated with thermal waters.



**Plate 3.** Representative scheme of fluid circulation at Stromboli. Most of the volatiles (around 90% of the total mass) are released through the open-conduit degassing, with a gas output of approximately of  $3 \times 10^6 \text{ Mg year}^{-1}$  [Allard *et al.*, 1994; Allard *et al.*, this volume]. Rising fluids give origin to high-temperature fumaroles (up to  $410^\circ\text{C}$ ), located close to active craters, low-temperature fumaroles (as the SC5 site) located in the Pizzo area, and anomalous  $\text{CO}_2$  fluxes from soil at the summit.

More than 90% of the gas is released from the open-conduit degassing system. The total output of gas emitted from the plume was estimated to be of the order of  $10^6$  Mg year<sup>-1</sup> [Allard *et al.*, 1994; Allard *et al.*, this volume].

Gas emitted both from high-temperature fumaroles (up to 410°C) and from low-temperature fumaroles located at the summit showed chemical and isotope signatures of an undoubted magmatic origin.

Anomalous CO<sub>2</sub> fluxes from soil were recognized both at the summit and in peripheral areas. Summit zones are characterized by extremely high fluxes due to gases being released laterally from the conduit (primary degassing). Diffusive gas emissions at low altitude probably derive from volatiles separated from boiling and/or degassing thermal aquifers (secondary degassing).

Thermal water originates from mixing between seawater and meteoric waters. The chemical composition of both of these end-members is modified from their pristine ones by leaching of host rocks and the dissolution of the CO<sub>2</sub>-rich gas phase as well.

Based on their chemical composition and the isotopic signature, volatile species dissolved in the thermal waters of Stromboli are clearly of magmatic origin and reveal intense gas–water interaction processes. Dissolved He and CO<sub>2</sub> contents, as well as the helium isotope ratios highlighted significant variations over time related to volcanic activity.

Such features highlight the scientific relevance of these peripheral manifestations, as suitable sites for monitoring the volcanic activity of Stromboli, especially during periods of intense Strombolian activity when summit areas are inaccessible.

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F. Grassa, S. Inguaggiato, and M. Liotta, Istituto Nazionale di Geofisica e Vulcanologia, Sezione di Palermo, Via Ugo La Malfa, 153-90146 Palermo, Italy.

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## Chapter 25 – AGU Calvari

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