

Preliminary geochemical and isotopic data on springs along the Fella-Sava fault zone (NE Italy)

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ABSTRACT During 2006, a geochemical survey was carried out on three perennial springs discharging along a segment of the Fella-Sava line, a transpressive lineament running in the eastern Southern Alps (Friuli, NE Italy). Waters have both a Ca-bicarbonate and a CaMg(Na)-HCO₃SO₄ sulfureous character. Oxygen and hydrogen isotopic data indicate a meteoric origin from catchments with different mean topographic elevation. The Sr isotopic composition of the Ca-bicarbonate water is consistent with circulation in Middle Triassic limestones; sulfureous waters have ⁸⁷Sr/⁸⁶Sr isotopic ratios which indicate interactions with the Upper Permian dolomite-evaporite layers, the latter representing the preferential decollement horizons during the compressive Alpine cycle. The isotopic data, the negative redox potential and the tritium activity for sulfureous springs suggest a long-lasting flow through different fracture systems. Limited effects of mixing between the deeper and superficial, low-salinity waters are observed. Speciation calculations indicate that sulfureous waters are oversaturated in calcite and silicate minerals, suggesting that cement deposition is an active process along the flowpaths. The formations of diagenetic seals in a compressive regime would contribute to fluid overpressuring in pores at a relatively shallow depth, where part of the seismic activity is concentrated. The SO₄, Ca and Mg concentrations, in one of the sulfureous springs, show a marked increase which seems to be related to the seismic events in the area, and is attributed to the expulsion of pore fluids with high concentration of sulphate due to strain changes.

1. Introduction

A number of field observations suggest that an interconnected system of fluid-filled cracks and pore fluid honeycombs characterize the continental crust. In active tectonic settings, such as along reverse faults in compressive regimes, a directional flow of these fluids within the fault meshes and shear fractures may be promoted in response to pressure gradients (Sibson, 1990). At seismogenic depth, both fluids generated by mineral dehydration reactions or meteoric waters, carried downward through the country rocks and channelized into the host fault zone, play a key role in determining the effective normal stress and in controlling the strength of crustal faults (e.g. Hickman *et al.*, 1995). In particular, the increasing fluid pressure, confined to small cracks and pores in the fault gauge, reduces all the normal stresses within a rock mass in the interseismic period (e.g. Sibson, 1974; Byerlee, 1992).

Waters migrating through a fault change their chemistry through reactions with rocks and minerals (e.g. Toutain and Barbon, 1999; Pizzino *et al.*, 2004). Water flow is mostly controlled by the interplay between the gauge and country rocks permeability which evolves with progressive deformation (Rojstaczer *et al.*, 1995). The hydrochemical variations observed in groundwaters prior or during a seismic sequence may hence be attributed to the effects of interseismic and coseismic strain changes, which include pore fluid release and mixing of different aquifers (e.g. Valenza and Nuccio, 1993; Hartmann and Levy, 2005), representative of crustal transients related to the mechanical behaviour of the crust (Bernard, 2001; Heinicke *et al.*, 2006).

A model of fluid circulation, including recharge areas, the mixing of water reservoirs at different depths, the influence of seasonal changes and the assessment of the geochemical background of the aquifer, is necessary before attributing hydrochemistry changes to a seismogenic structure.

In the present study, preliminary data concerning the solute geochemistry and the O, H, Sr isotopic composition of groundwaters sampled in an area running along a segment of the Fella-Sava (FS) line are reported. The FS fault represents a seismogenic structure related to the nealpine evolution of the eastern sector of the Southern Alps in Friuli (NE Italy), limiting the inner part of the Julian Alps to the north (Merlini *et al.*, 2002).

2. The Fella-Sava line: geological outlines and seismicity

The FS line is located in the eastern portion of the Southern Alpine chain (Fig. 1a), in an area which represents the retrobelt of the Alpine subduction. This area underwent several cycles of deformation resulting from the suture of Europe due to the motion of the African plate, yielding prevalently compressive structures and fragmentation. This complex structural evolution is evidenced by the remnants of structures related to the Hercynian orogeny, which outcrop mostly north of the Gail line in the Austro-Alpine domain, but also found in the northernmost sector of the Southern Alps. The east-westward strike and south-verging overthrusts which characterize this sector of the chain are attributed to the compressive Alpine phase (e.g. Carulli and Ponton, 1992). In particular, the external part of the chain is characterized by south-verging embriated structures in part reactivated during the Alpine orogeny, while the inner part of the chain is characterized by a sinclinorium (Merlini *et al.*, 2002) which is truncated to the north by the FS line. The FS line is a high-angle reverse fault (Venturini, 1990) which juxtaposes Permo-Scitic Units constituted by carbonatic and silico-clastic deposits (Werfen Formation) and evaporitic sequences (Bellerophon Formation) outcropping to the south and the Mesozoic carbonate terranes composed by dolomitic limestones outcropping to the north (Fig. 1c).

The area under study is characterized by intense seismic activity (Carulli *et al.*, 1990). In particular, the FS fault separates a northernmost part of the Southern Alps which is seismically inactive from the most active southernmost domains, acting as a mechanical disengagement (Slejko *et al.*, 1989), and it is interpreted as a transpressive dextral, potentially active, seismic lineament in the eastern Southern Alps (Doglioni, 2000).

The historical seismicity of the region is poorly documented because there were no important villages in that area of the mountains. The strongest event reported in the Italian earthquake catalogue

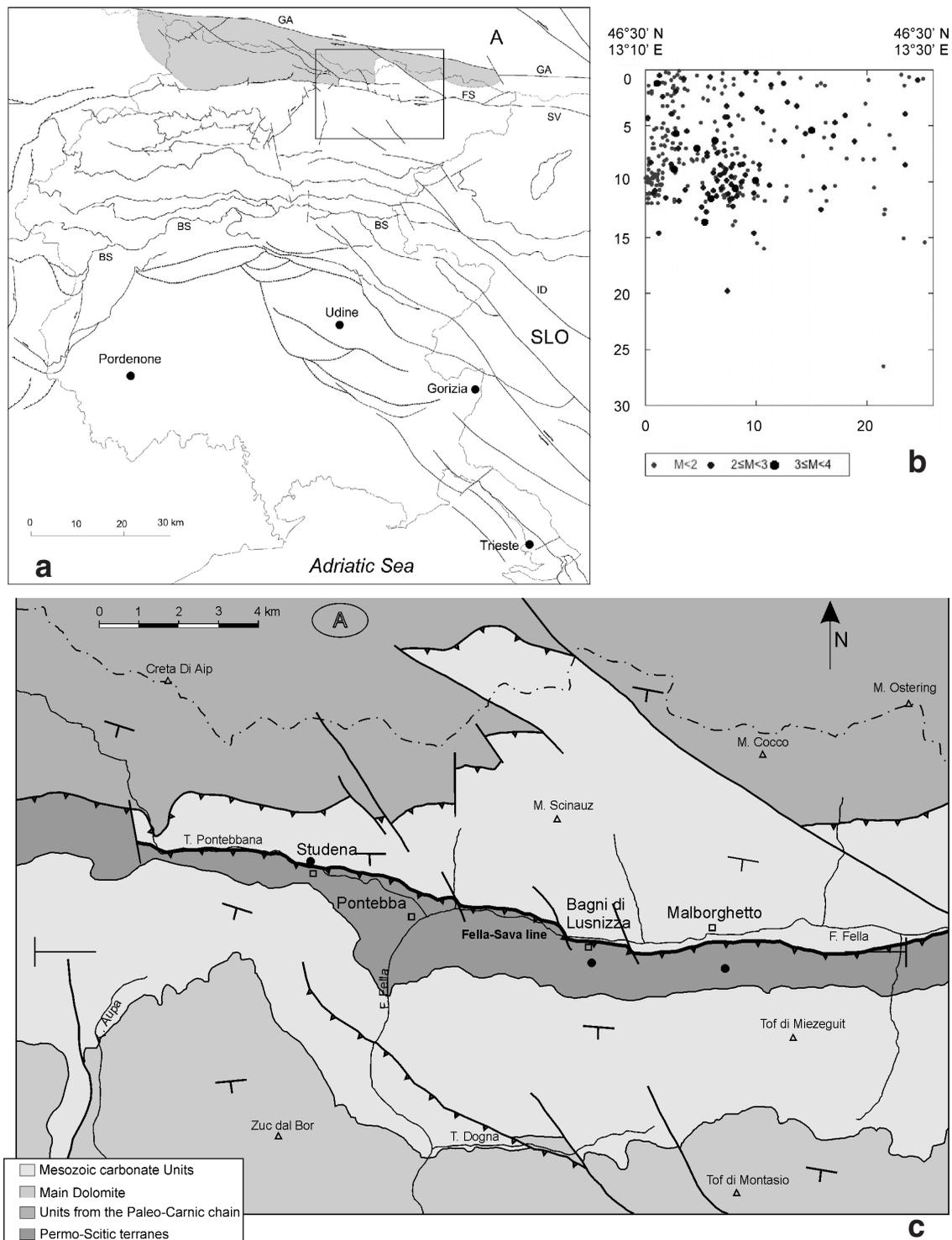


Fig. 1 - a) Map of the principal structural frameworks of the region. The Fella-Sava (FS) that becomes the Sava line (SV) in Slovenia, Barcis-Staro Selo (BS), Gail (GA) and Idrija (ID) faults are labeled. The box encompasses the studied segment of the Fella-Sava fault. b) Vertical cross-section, 10 km wide, across the studied segment of the Fella-Sava fault with earthquake hypocentres recorded since 1977 (from the OGS seismometric network). c) Schematic geological map of the studied area. Filled circles show water sample locations and open squares show town locations.

(Gruppo di lavoro CPTI, 2004) is that of July 10, 1908 which reached an epicentral intensity between VII and VIII MCS. Doubts remain about the strong earthquake of March 26, 1348, known as the Villach earthquake because very severe damage was reported in that town. Recent investigations (Slejko, 2006) suggest that the epicenter could have been located in the north-eastern sector of Friuli and, consequently, in the study area. The seismicity related to the studied part of the FS fault since the Seventies is shown in Fig. 1b. In particular, it can be observed that the hypocentral distribution in the period 1977-2006 is characterized by a maximum of foci at shallow depth (less than 3 km) and between 7-12 km, and that the number of events sharply decreases at depths greater than 13 km. This observation supports the hypothesis that the deepest segments of the crust underwent mostly aseismic deformation (Doglioni, 2000). Furthermore, the analysis of seismic data reveals that most of the earthquakes have low magnitude ($M < 3$).

The area is characterized by an intense ground- and surface water circulation. The hydrological catchments are delineated by E-W and N-S trending valleys, which represent the morphological expressions of the FS line and other minor lineaments, where the Fella river and tributaries flow. Among the springs discharging in proximity of the FS line, three, being perennial (the Malborghetto, Lusnizza and Studena springs), were suitable for geochemical monitoring. These waters were sampled monthly during a preliminary survey (from March 2006 to December 2006) and analyzed in order to evaluate composition and behavior of possible geodynamic relevance.

3. Analytical procedures

Eh, pH, temperature and electrical conductivity were measured directly in the field; HCO_3^- was measured by titration with 0.1 N HCl. Samples were stored in clean polyethylene bottles rinsed with the same water that was sampled. Major cations and trace elements were measured on samples filtered at 45 μm and stabilized by ultrapure HNO_3 for cation analysis. Major constituents were determined by a Dionex DX 120 ion chromatograph, and trace elements by a Jobin Yvon ICP-OES Ultima Due. $2\text{-}\sigma$ errors are within 2 and 5%, respectively.

H and O isotopic determinations on water samples were performed by equilibration technique for oxygen and water reduction (hydrogen production by using granular Zn). Measurements were carried out using a Finnigan Delta Plus mass spectrometer and AP 2003 automatic preparation system coupled with an IRMS. Analytical precision is better than 0.2‰ for $\delta^{18}\text{O}$ and 1‰ for δD .

The Sr isotopic compositions on solute species were obtained on the filtered samples, after Sr collection, using standard chromatographic methods and loading samples on single tungsten filaments, using Ta-chloride as an emitter. Isotopic analyses were carried out by a VG 54 mass-spectrometer. The measured $^{87}\text{Sr}/^{86}\text{Sr}$ ratios were fractionation-corrected to $^{87}\text{Sr}/^{86}\text{Sr}=0.1194$; data acquisition and reduction followed the procedure of "Analyst" (Ludwig, 1994). The experimental uncertainties on Sr isotopic ratio represent in-run statistics at 95% confidence level.

4. Water geochemistry

The major anion and cation concentrations measured in waters and some physico-chemical parameters are given in Table 1. Trace element abundances, limited to the May, 2006 sampling,

Table 1 - Concentrations of the major dissolved elements (mg/l) during the survey. nd = not determined.

Spring	date	T (°C)	pH	Cond	Eh	Ca	Mg	Na	K	Cl	HCO ₃ ⁻	SO ₄ ²⁻
Lusnizza												
46°29.714' N 13°22.705' E	03/15/06	8.9	7.2	3210	-274.7	413.22	87.25	4.30	2.38	1.22	231.80	1132.55
	05/07/06	8.9	7.4	2220	-273.8	351.70	80.69	2.27	0.95	3.79	210.45	1010.55
	06/27/06	10.1	7.4	1950	-232.7	457.71	88.96	3.47	0.77	4.11	219.60	1272.80
	07/31/06	10.1	7.3	1969	-280.3	444.69	89.08	3.56	0.81	nd	207.40	1231.97
	09/04/06	9.7	7.3	1905	-279.6	408.42	89.08	3.08	1.07	nd	219.60	1160.40
	10/17/06	8.5	7.4	2060	-243.1	434.07	88.47	2.30	1.33	1.77	115.90	1194.99
	12/11/06	7.7	7.5	1825	-262.7	396.39	81.42	1.49	0.90	nd	198.00	1074.91
Malborghetto												
46°29.641' N 13°26.615' E	03/31/06	10.6	7.3	1610	-92.0	308.42	82.64	4.21	2.26	7.30	213.50	886.63
	05/07/06	10.8	7.3	1616	-92.0	341.48	87.62	3.22	1.80	7.30	213.50	965.88
	06/27/06	10.7	7.2	1368	-65.3	354.31	91.75	1.31	1.27	3.35	231.80	1009.59
	07/31/06	10.6	7.1	1555	-95.5	323.65	83.85	4.02	1.37	nd	222.65	941.39
	09/04/06	9.0	7.3	1559	-127.7	352.10	91.51	3.17	2.61	nd	237.90	1016.31
	10/17/06	8.0	7.5	1563	-108.0	309.02	81.66	1.38	2.74	nd	234.85	879.91
	12/11/06	8.3	7.4	1573	-133.2	301.20	78.75	2.53	1.41	2.91	128.10	905.85
Studena												
46°31.301' N	03/15/06	9.7	8.7	475	173.4	69.94	20.42	1.17	1.85	2.89	283.65	23.34
13°16.030' E	12/11/06	10.0	7.3	398	148.4	58.32	17.01	1.82	1.80	2.29	234.85	15.85

are reported in Table 2. The waters are cold, nevertheless their temperature remains quite constant throughout the survey, being much higher than the ambient surface temperature during winter.

The major chemical constituents allow us to distinguish a Ca-HCO₃ water (Studena) from CaMg(Na)-HCO₃(Cl)SO₄ sulfureous waters (Lusnizza and Malborghetto). The sulfureous waters have the electrical conductivity of mature waters with negative redox potential (Eh), suggesting reducing environments. The Eh levels, coupled with pH values, indicate that the Studena groundwater falls between the M_nO₂/Mn²⁺ and Fe(OH)₃/Fe²⁺ buffers, while at Malborghetto and Lusnizza they are buffered by sulfate reduction likely due to bacterial activity. Since bacterially mediated reactions that tend to lower the Eh are slow in ancient rocks, it can be inferred that the sulfureous waters of Malborghetto and Lusnizza have quite long residence times in the aquifer.

The Ca/SO₄ molar ratio close to unit suggests gypsum or anhydrite leaching; the Mg-SO₄ correlation observed for the Malborghetto spring (not shown) indicates that in this case dissolution of Mg-sulfate minerals also played a significant role. The Na/Cl molar ratio greater than 1 in some of the samples is interpreted as the possible evidence of the influence of clay minerals such as Na-rich smectites or interaction with Na-silicate on water chemistry, which has however negligible effects on the Ca²⁺ budget (Boschetti *et al.*, 2005).

The Lusnizza sulfureous waters have significantly higher Sr concentration with respect to the

Table 2 - Minor and trace element concentrations in $\mu\text{g/l}$ (Si in mg/l).

	Lusnizza	Malborghetto
Si	2.51	2.38
Li	5.62	9.24
B	76.8	27.6
Al	0.5	0.3
Mn	<0.1	2.07
Fe	0.94	122.70
Co	0.52	0.58
Ni	1.28	0.15
Zn	1.23	1.55
Ga	0.11	0.10
As	<0.1	60.17
Rb	0.94	3.82
Sr	8298	6061
Mo	2.48	4.57
Cs	<0.1	1.09
Ba	14.1	10.3
U	0.27	0.90

Malborghetto ones, possibly due to a higher extent of anhydrite and gypsum dissolution, which accounts for a different circulation. The relatively higher Rb content with respect to low-salinity waters supports the involvement of micaceous minerals.

The WATEQ4F geochemical code (Ball and Nordstrom, 1991) was used to calculate activity coefficients of species in solutions and the saturation index ($SI = \log IAP/KT$, where K is the equilibrium constant associated to dissolution reactions, T the temperature and IAP the ion activity product). Sulfureous waters are invariably oversaturated in calcite [Lusnizza: average (av.) $SI = 0.29 \pm 0.09$; Malborghetto: av. $SI = 0.16 \pm 0.10$] and quartz ($SI = 0.22$ and 0.37 for Lusnizza and Malborghetto, respectively), the latter, possibly forming from amorphous silica dissolution. Waters are also oversaturated in mineral phases associated to alteration of silicates (kaolinite and related minerals) and in micaceous and clay minerals possibly derived from hydrolysis of feldspar (K-mica, pyrophyllite,

beidellite), and undersaturated in disordered dolomite (Lusnizza: av. $SI = -0.67 \pm 0.2$; Malborghetto: av. $SI = -0.75 \pm 0.26$), anhydrite (Lusnizza: av. $SI = -0.45 \pm 0.05$; Malborghetto: av. $SI = -0.59 \pm 0.04$) and gypsum (Lusnizza: av. $SI = -0.20 \pm 0.04$; Malborghetto: av. $SI = -0.34 \pm 0.03$). These data suggest that waters are able to dissolve evaporite-related minerals and to precipitate calcite and other phases related to the contribution of clayed rocks, reducing rock permeability and placing the conditions for sealed compartments to form.

5. O-H-Sr isotopes

The O-H-Sr isotopic data are given in Table 3. Oxygen and hydrogen isotope ratios in the sampled waters range between -7.9 to -10.8 for $\delta^{18}\text{O}$ (‰ vs. SMOW) and between -51 to -72 for δD (‰ vs. SMOW).

In the δD vs. $\delta^{18}\text{O}$ diagram (Fig. 2) springs are isotopically distinct and plot close to the linear regression for northern Italy precipitations (Longinelli and Selmo, 2003) indicating a meteoric origin from catchments with different mean topographic elevation. Seasonal variations are not observed.

^3H analysis on the Lusnizza and Malborghetto groundwaters yielded values of 11.8 and 8.7 TU, respectively. In the well-mixed reservoir approach (Michel, 2005) the observed tritium concentrations are reproduced by waters that were isolated from the atmospheric recharge for over 40 years, suggesting a long-term circulation.

Table 3 - $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and oxygen- and hydrogen-isotope results.

Spring	$^{87}\text{Sr}/^{86}\text{Sr}$	2- σ	$\delta^{18}\text{O}$	δD
Lusnizza				
03/15/06	0.707428	0.000021	-9.8	-66
05/07/06	0.707425	0.000022	-9.9	-65
06/27/06	0.707437	0.000019	-9.8	-66
07/31/06	0.707442	0.000020	-9.8	-64
09/04/06	0.707451	0.000021	-9.5	-65
10/16/06	0.707401	0.000020	-10.1	-63
12/11/06	0.707432	0.000022	-9.9	-66
Malborghetto				
03/31/06	0.707515	0.000022	-10.3	-69
05/08/06	0.707481	0.000020	-10.3	-70
06/27/06	0.707509	0.000021	-10.4	-72
07/31/06	0.707504	0.000019	-10.4	-69
09/04/06	0.707481	0.000020	-10.5	-71
10/16/06	0.707492	0.000022	-10.7	-70
12/11/06	0.707526	0.000022	-10.8	-69
Studena				
03/15/06	0.707796	0.000021	-7.9	-52
12/11/06	0.707839	0.000044	-7.9	-51

The Sr isotopic composition ranges between 0.707401 – 0.707451; 0.707481 – 0.707526 and 0.707796 – 0.707839 for Lusnizza, Malborghetto and Studena springs, respectively. It has been observed that sulfurous waters did not show significant seasonal changes during the surveyed period (Fig. 3) and that Malborghetto has higher $^{87}\text{Sr}/^{86}\text{Sr}$ than Lusnizza, which is coupled with the higher Rb/Sr ratio measured in the May 2006 sample. This could indicate a more advanced dissolution with involvement of micaceous minerals.

The Sr isotopic composition in groundwaters reflects the water-rock interaction; moreover, the degree to which the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio approaches the dissolving Sr depends on velocity, infiltration flux, aquifer thickness and weathering rates (e.g. Johnson and DePaolo, 1994). The Sr isotopic composition of Studena is that expected for an aquifer confined into Middle Triassic limestones (Howarth and McArthur, 1999) suggesting dominant carbonate dissolution, while the correlation between $^{87}\text{Sr}/^{86}\text{Sr}$ ratio and sulfate content observed for Malborghetto and Lusnizza indicate that the dominant sources of non-radiogenic Sr in waters are gypsum and anhydrite from the Bellerophon Formation (Permian) (Fig. 4).

The combined Sr and O-H isotopic data suggest the drainage from distinct hydrological

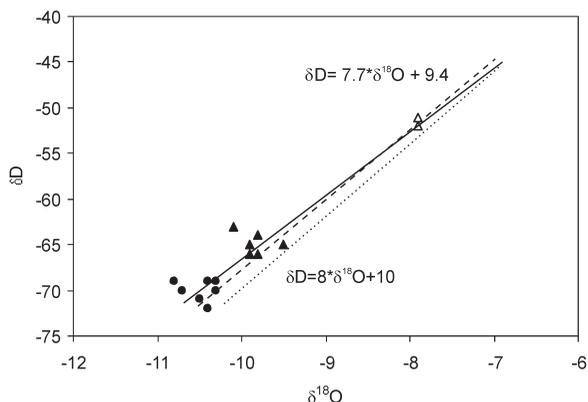


Fig. 2 - Hydrogen and oxygen isotopic composition of Lusnizza (solid triangle), Malborghetto (solid circle) and Studena (open triangle) springs. The dashed line refers to precipitation of northern Italy (Longinelli and Selmo, 2003); the dotted line is the global meteoric water line (Craig, 1961) and the solid line represents the linear regression for the studied samples.

circuits for the different springs. Furthermore, a correlation between $\delta^{18}\text{O}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ is observed for Lusnizza (Fig. 5), indicating that the mixing of different aquifers could account for the small variations observed in the isotopic compositions. The hypothesis of mixing is also supported by the positive correlation between the Na content and the temperature (not shown), interpretable as due to dilution with shallow, low-salinity waters possibly confined in a karst circulation in the overlying carbonates.

6. Temporal variations and seismic activity

The hydro-geochemical data were collected on average each month, so that co-seismic effects cannot be unequivocally revealed. Nevertheless, temporal variations in some chemical species have been observed at the Malborghetto and Lusnizza springs. In particular, the Malborghetto groundwater shows an increase in the Ca, Mg and SO_4 content (Fig. 6) in association with the seismic event recorded on June 24, 2006 ($M= 2.2$, depth 4.0 km) with epicenters located at

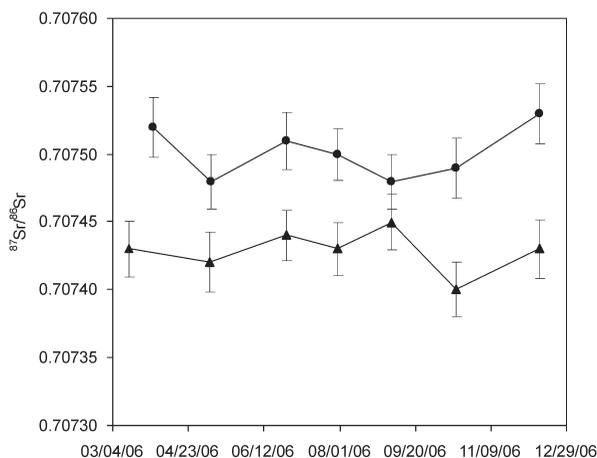


Fig. 3 - Temporal trends of $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratio for the Lusnizza and Malborghetto sulfureous springs. Symbols as in Fig. 2.

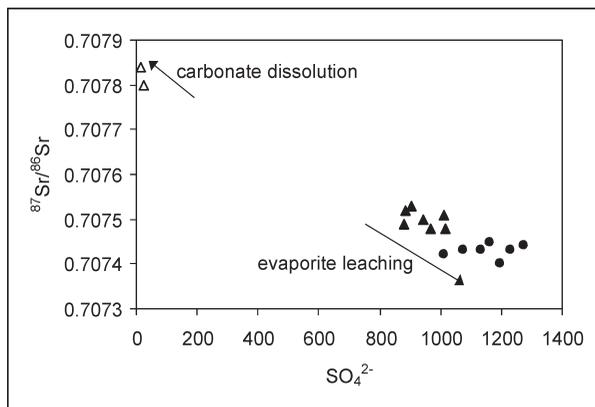


Fig. 4 - ⁸⁷Sr/⁸⁶Sr ratio vs. sulfate content (ppm) for the Malborghetto, Lusnizza and Studena springs. Symbols as in Fig. 2.

46.574° N, 13. 250° E, about 10 km west of the spring, and that recorded on September 12, 2006 (M=2.7, depth 5.6 km) with epicenter at 46.470° N, 13.812° E, about 30 km east, which represent the only seismic activity recorded in the area during the investigated period. An increase of Ca and SO₄ concentration, even if less significant, is also recorded by the Lusnizza groundwater. The observed chemical variations are attributed to the expulsion of pore fluids with high sulfate concentration into the circulating groundwaters, due to pore collapses related to strain changes. Similar trends of increasing sulfate concentration both co- or post-seismic were reported by Favara *et al.* (2001) for the 1968 Belice (western Sicily) earthquake (M=6.1); Biagi *et al.* (2006) for the 1992 Kamchatka earthquake (M=7.1) and Nishizawa *et al.* (1998) for the relatively low-magnitude (M<3, M<4.8) earthquake swarms of the Izu Peninsula (Japan). A pre-seismic increase in sulfate concentration is documented by Song *et al.* (2003) for the 1999 Chi-Chi earthquake (M=7.6) in Taiwan.

The lack of hydrogeochemical signals due to the input of deep-seated fluids, such as a change in the ⁸⁷Sr/⁸⁶Sr ratio towards values equilibrated in the crystalline basement, suggests that the root of the sismogenic FS fault segment was probably confined in the overlying sedimentary

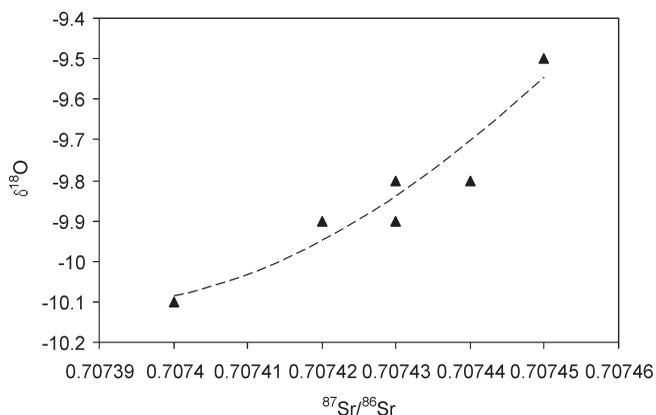


Fig. 5 - δ¹⁸O vs. ⁸⁷Sr/⁸⁶Sr for the Malborghetto spring, showing a possible mixing line.

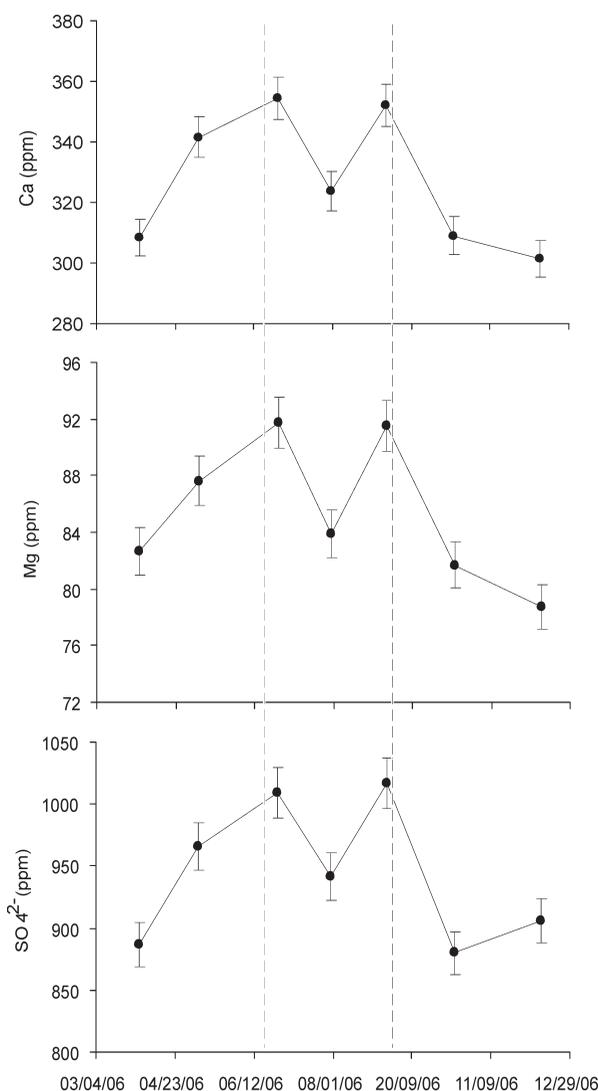


Fig. 6 - Variations of concentration values for Ca, Mg and SO₄²⁻ vs. time in the Malborghetto spring. The dashed vertical lines indicate the occurrence of the June 24, 2006 and September 12, 2006 earthquakes.

sequence. This is consistent with the estimated, relatively shallow depth of the seismogenic structures (Fig. 1b). It is hence, tentatively suggested that the Malborghetto aquifer is confined to structural zones that respond to stress increment, and could be the site for further surveys.

7. Conclusive remarks

Water chemistry and isotope data indicate that sulfurous groundwaters do not lie along the same evolutionary flowpath, suggesting the occurrence of distinct reservoirs in different fracture systems, possibly extending on either side of the main fault zone as found in steep reverse faults.

Speciation calculations suggest that calcite, quartz and other precipitate depositions are an

active process along the flowpath, promoting cracks and sealing open veins. It is generally accepted that the rate of fracture sealing is rapid with respect to geodynamic rates (e.g. Blanpied *et al.*, 1992), and the observation that fluids are still channelized into the FS fault zone suggests that the permeability decrease due to the filling of the fracture network by cement deposition is not fast enough with respect to the rate of fracture openings due to the recurrence of earthquakes.

Despite the short interval of sampling, the Malborghetto and Lusnizza springs show hydrochemical changes which seem to be the response to local seismicity, even if of low magnitude. These are interpreted as being the consequence of a fault-valve effect that causes an intermittent increase of SO₄, Ca and Mg due to temporary changes in structural permeability.

Additional data would be, however, necessary for a more accurate definition of the background variations, allowing for a better assessment of the significance of the observed anomalies.

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