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Pre- and syn-eruptive geochemistry of volcanic gases from Soufriere Hills of Montserrat, West Indies

G. Hammouya¹, P. Allard², P. Jean-Baptiste², F. Parello³, M.P. Semet⁴ and S.R. Young⁵

Abstract. Soufriere Hills fumaroles contained magma-derived volatiles before and during the eruption initiated in 1995 but also preserved a typical and quite steady hydrothermal composition. Chemical changes due to increased boiling and a greater input of oxidizing magmatic gas occurred only at Galway's Soufriere, the most active fumarolic field. Hydrothermal buffering of the fumaroles has been favoured by their remote location (1-2 km) from the eruptive vents and by a preferential degassing of the uprising magma through intrusive conduits under the crater. High temperature (720°C) gas collected from the extruding lava dome in Feb. 1996 was chemically and isotopically representative of the magmatic gas stream. Its composition allows assessment of average eruptive fluxes of H₂O, CO₂ and HCl which require the degassing of only 2.5-3 times more magma than erupted.

Introduction

Over its last centuries of dormancy Soufriere Hills (SFH) volcano has maintained an intense hydrothermal activity, with hundreds of steaming vents, mud pools and hot water discharges extending at 310-570 m a.s.l on its outer flanks, within four main fields: Galway's (GS), Gages Upper (GUS) and Lower (GLS), and Tar River (TRS) 'Soufrieres' (Fig. 1). These manifestations, linked to a high-enthalpy ($\approx 250^\circ\text{C}$) hydrothermal system [Chiodini *et al.*, 1996], increased noticeably during intrusion-related seismic crises in 1933-37 [Perret, 1939], 1966-67 [Shepherd *et al.*, 1971] and, then, before the current eruption. No manifestation existed inside the summit crater before phreatic vents suddenly opened there in July-August 1995, giving rise to extrusion of an andesitic lava dome since mid-November 1995 [Young *et al.*, 1998].

Here we report and discuss chemical and isotopic data for SFH fumaroles, that were obtained before (up to 30 years ago) and during the current eruption, allowing assessment of precursory signals of the events. We also report unique results for a 720°C gas collected from the extruding lava dome. These help to characterize the local magmatic source and constitute the highest temperature datum ever obtained for a volcanic fluid in the Lesser Antilles Arc.

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Geochemical Evolution of Fumaroles

SFH fumaroles were early studied by Perret (1939), but their first quantitative analysis dates from the 1966-67 seismic crisis (M. Chaigneau, unpub. data; Table). Sparse data were further gathered in 1985-1989 [IPG Observ. Guadeloupe, unpub.] and, for geothermal exploration, in 1991-92 [Chiodini *et al.*, 1996]. Stimulated by increasing pre-eruptive seismicity, a fumarole survey was started in March 1995 and was then intensified after the eruption onset [Hammouya *et al.*, 1996], through a collaboration between MVO (Montserrat Volcano Observatory) and the Soufriere Observatory in Guadeloupe (where routine chemical analysis was operated). This survey lasted until pyroclastic flows threatened the fumarolic areas, all of which were progressively buried or destroyed. This enhances the interest of the results obtained, despite their sparsity and/or incompleteness for some species (H₂O, CO and He). The largest dataset is for Galway's Soufriere, the most accessible but most active field. More discrete results were obtained at other sites. The Table reports analyses for each field at representative dates, together with isotopic results for He and C. The following features can be outlined:

(1) Over the whole period investigated SFH fumaroles kept steady surface boiling temperatures of 96-98°C and a typical hydrothermal chemistry. H₂O (91-97%), CO₂ and H₂S were the dominant species, coexisting with minor amounts of H₂, CH₄ and CO. Air contamination was broadly low. H₂S was always very abundant, but SO₂ and HCl, which are typical components of magmatic gases, were never detected.

(2) He and C isotopic data obtained before (since 1992) and during the eruption however demonstrate a MORB-type magmatic derivation of helium and carbon dioxide emitted from GS, TRS and, to a lesser extent, GLS (Table). Particularly, both in March 1995 and February 1996 GS fumaroles had

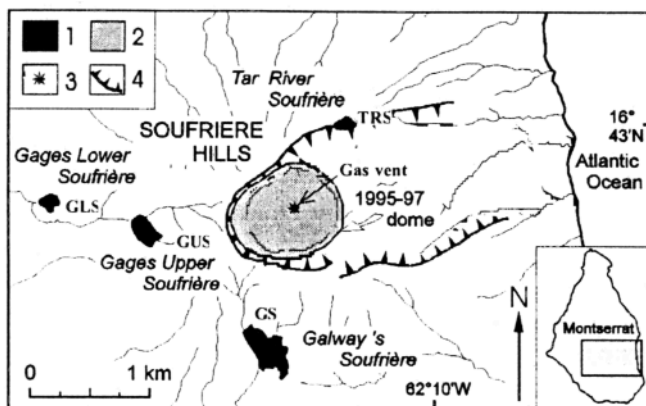


Figure 1. Map of Soufriere Hills showing 1: the fumarolic fields; 2: the 1995-97 lava dome, covering former Castle Peak dome (dashed contour); 3: the site of dome gas sampling (Feb. 1996); and 4: English's Crater rim.

Table. Chemical and Isotopic Composition of Fumarolic and Magmatic fluids of Soufriere Hills Volcano, Montserrat

Site	Date	T°C	H ₂ O % tot.	CO ₂ %	H ₂ S %	SO ₂ %	H ₂ %	CH ₄ %	CO ppm	N ₂ %	O ₂ %	He ppm	HCl %	S/C	³ He/ ⁴ He R/R _a	δ ¹³ C ‰
Galways ¹	15/07/66	98		72.8	25.6	0	0.02	0.04	0	2.5			0	0.35		
Galways ¹	23/09/67	98		70.0	28.9	0	0.20	0.10	0	0.6			0	0.41		
Galways ²	03/91	98	92.9	74.1	25.3	0	0.21	0.024	2.2	0.9		5.9	0	0.34		
Galways ²	09/92	98	92.2	71.7	27.4	0	0.096	0.037	0.4	0.4		6.3	0	0.38	8.2 (8.2)	
Galways	16/03/95	98	93.4	69.5	30.0	0	0.005	0.02		0.6	0.13	16.7	0	0.43	7.9 (7.9)	-5.6
Galways	11/07/95	98.4		68.4	30.3	0	0.012	0.012	0	1.1	0.22		0	0.44		
Galways	02/08/95	98.0		64.4	33.6	0	0.004	0.015	0	1.7	0.34		0	0.52		
Galways	22/11/95	96.2		62.2	36.0	0	0.006	0.039	0	1.4	0.32		0	0.58		
Galways	27/02/96	96.7	91.1	60.7	37.9	0	0.013	0.043	7.3	1.1	0.25	24.6	0	0.62	7.5 (7.5)	-5.3
Galways	04/06/96	98.0		58.8	39.6	0	0.045	0.054	0	1.2	0.39		0	0.67		
Tar River	27/05/89	98		82.5	15.8	0	0.08	0.20	0	1.2	0.26		0	0.19		
Tar River ²	03/91	98	96.6	80.3	17.6	0	0.091	0.08	0.6	1.3		7.3	0	0.22		
Tar River	16/03/95	97.1	95.5	79.4	16.2	0	0.082	0.09	1.2	3.4	0.89	9.0	0	0.20	8.8 (8.9)	-4.0
Tar River	12/07/95	97.2		77.3	19.9	0	0.085	0.08	0	2.2	0.44		0	0.26		
Tar River	14/11/95	96.7		77.9	19.3	0	0.08	0.11	0	2.2	0.48		0	0.25		
Tar River	28/02/96	98.0	95.9	79.2	17.3	0	0.092	0.11	0	2.8	0.65	29.8	0	0.22	7.9 (7.9)	-4.5
Upper Gages ¹	23/09/67	98		80.6	16.1	0	0.60	0.30	2000 [§]	2.1			0	0.20		
Upper Gages ²	03/91	98	95.6	87.0	11.6	0	0.49	0.43	0.6	1.3		4.5	0	0.13		
Upper Gages ²	09/92	98	96.5	86.6	11.1	0	0.43	0.43	0.5	0.7		4.3	0	0.13		
Upper Gages	27/07/95	97.8		83.0	14.6	0	0.35	0.31	0	1.4	0.33		0	0.17		
Lower Gages ²	03/91	99	95.2	91.2	7.7	0	0.10	0.62	0.6	0.8		6.2	0	0.08		
Lower Gages ²	09/92	99	96.4	91.9	6.9	0	0.11	0.51	0.4	0.9		6.1	0	0.07	5.9 (5.9)	
Lower Gages	27/02/96	96.4	96.0	83.2	11.3	0	0.096	0.55	0	3.9	0.90	16.4	0	0.13	6.3 (6.4)	-3.1
Lower Gages	07/04/97	97		66.0	7.6	0	0.09	0.36	0	19.8	5.5		0	0.11		
Lava dome [§]	27/02/96	720	90.3 [†]	19.4	3.8(S)	4.11	0.19	647	46.7	9.6	4.2	16.7	0.26	2.6 (7.7)		
Lava dome	27/02/96	720	91.8 [†]	15.1	0	3.64	0.49	598	65.5	15.1	<5					-5.9
Lava dome	27/02/96	720		1.8	0	0.5	1.29	0.18		76.8	19.5			0.28		
Lava dome edge	"	150														-5.2

H₂O in vol. % of total fluid, other components referred to the anhydrous gas phase (0 values: below detection; blanks: not determined). Data from: ¹M. Chaigneau (CNRS, unpub., quadrupole mass spectrometry); ²Chiodini et al., 1996 (gas chromatography); and this work (gas chromatography, plus acidimetric titration (CO₂) and electrochemistry (total S and Cl) in case of lava dome gas sampled through soda solution[§]). [†]H₂O without air contamination. [§]Uncertain value. Gas sampling in pre-evacuated glass bottles or/and in copper tubes (for He isotopes). ³He/⁴He ratios (±0.3%) are normalized to the atmospheric ratio (R_a = 1.38x10⁻⁶); values in brackets were corrected for air dilution based on sample's content in Ne [Chiodini et al., 1996] or N₂ (this work). δ¹³C values (±0.1‰) vs. PDB standard.

³He/⁴He ratio (7.5-7.9 R_a) and δ¹³C values (-5.3 to -5.6‰) very close to those found for the lava dome gas (7.7 R_a and -5.9‰). These isotopic data thus evidence a pre- and syn-eruptive magmatic contribution to SFH fumaroles, whose otherwise persistent hydrothermal signature requires the buffering by thermal aquifers. These data also suggest a magmatic origin of the abundant sulfur, SO₂ being readily converted to H₂S during water-gas-rock interactions [Giggenbach, 1987] and any associated HCl removed in aqueous solutions.

(3) Minor temporal variations were detected in GUS, GLS and TRS fumaroles (Fig. 2), whose compositional range can be ascribed to a different degree (or depth) of boiling of a single aquifer (curve in Fig. 2a) or interacting aquifers (Fig. 2b). In contrast, significant changes occurred in GS fumaroles, whose feeding source was clearly distinct. These changes included (Fig. 3): i) Smooth anticorrelated variations of S/C and H₂/CO₂ ratios from 1988 to mid-1995, with a tendency to higher sulfur and lower H₂ (also vs. CH₄) during pre-eruptive seismicity; ii) Sharp oscillations of the same ratios (with strong H₂ spikes, e.g. August 11, 1995) during the phreatic events; then iii) a marked increase of sulfur (S/C), CO and He (Table), but low H₂ during dome growth. A concomitant rise of He occurred at TRS and GLS, but with opposite trend in ³He/⁴He.

(4) Assuming a redox control of CH₄/CO/CO₂ ratios in the fumaroles by vapour-rock exchanges at depth [Giggenbach, 1987], we infer an equilibrium temperature of up to 272°C for GS fluids in 1996, compared to steady values of 161-201°C at

other sites (Fig. 4). Taking account of gas solubilities in water at such temperatures (H₂ ≈ CH₄ << CO₂ < H₂S), increased boiling beneath GS could simply account for the anticorrelated variations of S/C and H₂/CO₂ before the eruption. But this process alone cannot explain the large variations of H₂ vs. CH₄ before and during the phreatic events (Fig. 2b and 3), nor the synchronous rise of S/C and poorly soluble He and CO during dome growth. These trends suggest a greater inflow of SO₂-rich, comparatively oxidizing magmatic gas that affected the redox conditions of the aquifer, to which H₂ is much faster to adjust than CH₄ [Giggenbach, 1987].

Geochemistry of Lava Dome Gas

On February 27, 1996, just before the first pyroclastic flows, two of us (G.H. and P.A.) could briefly sample gas exhaled at 720°C from an incandescent fracture in the extruding lava dome (Table). This gas was strongly diluted and partly oxidized by air circulating among the lava blocks, but differed markedly from the fumaroles in containing SO₂, HCl and high amounts of H₂ and CO. Its CO/CO₂-H₂/H₂O ratios suggest an equilibrium oxygen fugacity close to both NNO and the H₂S-SO₂ buffers at 720°C (Fig. 4), somewhat lower than expected for a gas equilibrated with molten SFH andesite at 850°C (NNO+1(±0.5); [Barclay et al., 1998]). Such a shift is attributed to secondary alterations during our sampling, also responsible for excessive methane (see Fig. 4).

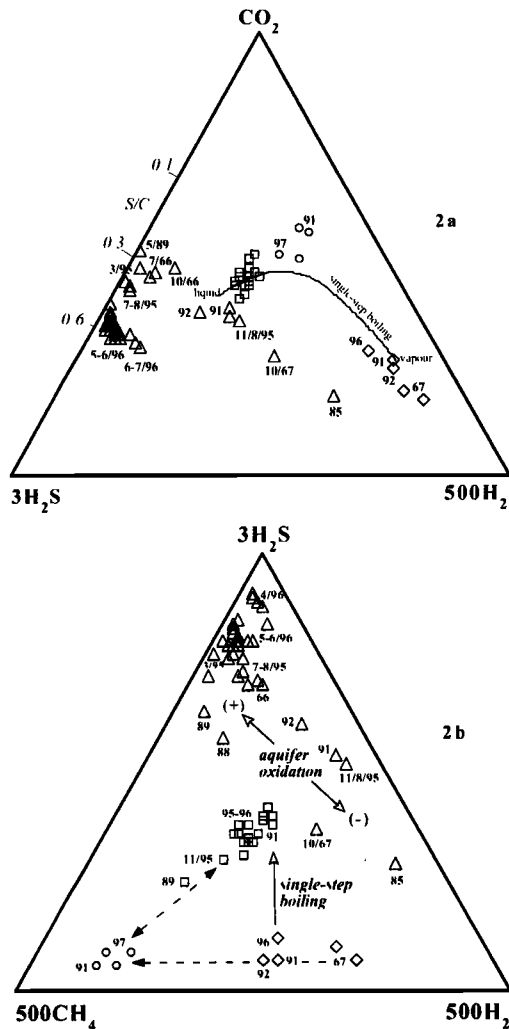


Figure 2. Triangular plots of CO₂-H₂S-H₂-CH₄ in Soufriere Hills fumaroles, 1966-1997: GS (triangles), TRS (squares), GUS (diamonds) and GLS (circles). **2a)** Curve: CO₂-H₂S-H₂ evolution during single-step boiling of a ~250°C aquifer with initial vapour phase similar to GUS samples [Chiodini *et al.*, 1996]. **2b)** GUS and TRS data points also defines mixing trends with a CH₄-rich GS-type fluid, at constant H₂/H₂S ratio. Owing to similar solubility behaviour of H₂ and CH₄, the large variations of H₂/CH₄ at GS cannot be due to boiling and, rather, suggest changing redox conditions in the aquifer, due to the input of more oxidizing magmatic gas. See text.

The high water content of the dome gas is typical of andesitic magmatic fluids in subduction zones [e.g. Allard, 1983]. In contrast, its greater richness in chlorine than sulfur (Cl/S≈4) is a rarer feature, encountered at a few arc volcanoes in post-eruptive rather than eruptive stage (e.g. Showa-Shinzan, in Japan; [Symonds *et al.*, 1996]). A high Cl/S ratio was however measured both in the volcanic plume [Allen, 1996; Oppenheimer *et al.*, 1998] and in crystal melt inclusions of SFH andesite [Hammouya *et al.*, 1996; Young *et al.*, 1998] and is thus characteristic of eruptive degassing at Soufriere Hills.

An average SO₂ flux of ca. 400 tons per day, equivalent to ca. 0.3 kg of S per ton of extruded magma (0.03 wt %), was emitted during dome growth in 1995-1997 [Young *et al.*, 1998]. Combining these numbers with the dome gas composition allows us to assess contemporaneous fluxes of ca. 900, 1400 and 25000 t.d⁻¹ of HCl, CO₂ and H₂O, respectively, supplied by a loss of 0.13, 0.21 and 3.7 wt % of

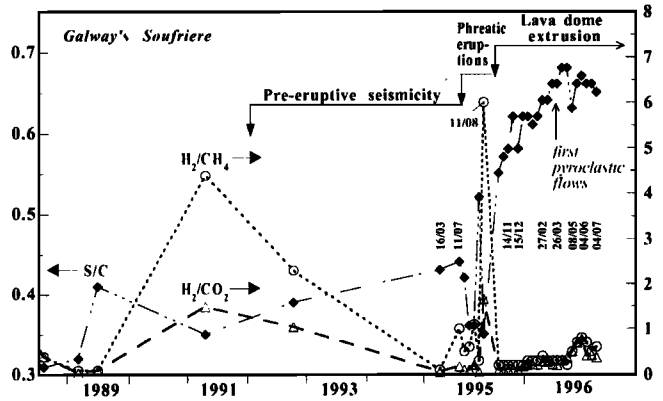


Figure 3. Pre- and syn-eruptive variations of S/C, H₂/CO₂ (x10¹) and H₂/CH₄ ratios in GS fumaroles (time scale x2 for 1995 and 1996). All analyses made with gas chromatography.

each same species from the extruded magma. The weight losses for HCl and H₂O compare fairly well with those derived from their respective abundances in crystal melt inclusions and degassed glass in the andesite (0.1-0.2 and 3.5-4.0 wt %; [Young *et al.*, 1998; Barclay *et al.*, 1998]). This supports a magmatic origin of H₂O and minor hydrothermal addition to the volcanic plume. Now, for an original melt fraction of 40% in the andesite [Devine *et al.*, 1998], the above volatile fluxes require the degassing of 2.5-3 times more magma than was erupted. Such a degassing excess, also verified for sulfur

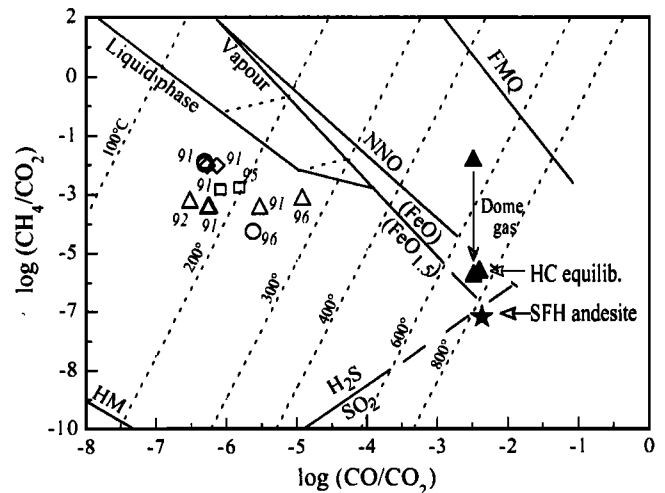


Figure 4. CH₄/CO₂ vs CO/CO₂ in SFH fumaroles, dissolved gas in GS thermal water (dotted circle) and lava dome gas (dark triangles), with respect to mineral and (H₂S-SO₂) redox buffers (modified from Giggenbach, 1987). FMQ: fayalite-magnetite-quartz; NNO: nickel-nickel oxide; HM: hematite-magnetite. Isotherms refer to redox equilibrium buffered by vapour-rock exchange. All fumaroles plot below the rock-buffered liquid-vapour domain typical for common hydrothermal systems, i.e. towards more oxidizing (magmatic) conditions, with inferred equilibrium temperatures ranging from 161°C (GLS) to 272°C (GS). 'HC': equilibrium composition of the dome gas at 720°C and log fO₂ = -15.77, as computed from its measured CO/CO₂ ratio (thermodynamic data from Giggenbach, 1987). Excessive methane (CH₄/CO₂ ratio) in samples is due to a pollution from burning of our silicone tight upon sampling. Star: theoretical plot for a gas equilibrated with SFH andesite magma in the P-T redox conditions of its reservoir (~NNO+1 at 850°C and P_{H₂O} of 110-130 MPa; Barclay *et al.*, 1998).

[Young *et al.*, 1998], is quite small compared to that observed at other erupting arc volcanoes [e.g. Andres *et al.*, 1991; Allard *et al.*, 1994]. It suggests a moderate gas saturation of SFH magma prior to eruptive ascent from its storage system.

Discussion and Conclusions

The data reported here and those for thermal waters in Montserrat [Chiodini *et al.*, 1996] strongly suggest that hydrothermal activity at Soufriere Hills has long been sustained by a deep supply of magmatic heat and gas to the overlying aquifers. This is supported by $^3\text{He}/^4\text{He}$ data (we also found a ratio of 5 R_a in gas bubbling through Hot Pond thermal springs, along the northern coast, in March 1995) and by the position of all SFH fumaroles in Figure 4; these plot below the rock ($\text{FeO-FeO}_{1.5}$) buffered vapour-liquid domain typical of most hydrothermal systems [Giggenbach, 1987], i.e. towards more oxidized (magmatic) redox conditions.

The growth of fumarolic activity during previous non-eruptive intrusions and before the current eruption points to an increase of this supply due to magma ascent and decompression. Now, the minor chemical changes in most fumaroles in 1995-96, their preservation of typical hydrothermal features, and the lack of any phreatic eruption (i.e. of excess gas loading) in all fumarolic areas imply that degassing of the uprising magma has preferentially been forced through central intrusive conduits under the crater. This is consistent with evidence of extensive sealing of the magma conduit walls [Boudon *et al.*, 1998] and with the peripheral confinement of all fumarolic manifestations at remote distance (1-2 km) from the eruptive vents (Fig. 1). Such conditions were favourable to hydrothermal buffering.

Fumaroles at Galway's Soufriere have been more sensitive to the magmatic input, even though their $^3\text{He}/^4\text{He}$ ratio was not the highest measured one. From 1967 to 1992 their composition (S/C ratio) remained quite unchanged (Fig. 2a), excepting a great variability in minor H_2 , partly due to uncertainty on pre-1988 analyses by mass spectrometry. Their evolution before and during the phreatic events in 1995 point to increasing disturbance of their feeding aquifer as the magma was approaching the surface, fracturing the volcanic pile. While gas-rich aqueous solutions could gain a temporary access to the central conduit zone before being blasted out at the crater (we measured 12 g/kg of Cl in a plume condensate from the July 28, 1995 phreatic eruption, compared to 26 g/kg in dome gas), more reduced hydrothermal fluids were occasionally entrained beneath GS, leading to the sharp H_2/CH_4 spikes superimposed on a longer trend of decreasing H_2 attributed to greater aquifer boiling and oxidation. Enhanced water-rock interactions and mixing with fluids richer in helium but poorer in ^3He is also suggested by the lowering of $^3\text{He}/^4\text{He}$ at both GS and TRS (note, however, that $^3\text{He}/^4\text{He}$ ratio at all sites gradually converged towards the dome gas ratio). The system reached a more steady regime when magma extrusion and degassing developed through the open vents, with a possible reversed evolution of S/C at GS in June 1996 (Fig. 3).

This work further outlines that, whenever possible, geochemical monitoring of comparable dormant volcanoes must be done on fumaroles the closest to magma conduits, where independent gas columns can rise through aquifers and, thus, bring less filtered indication of eruptive potential.

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