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## Gases and helium isotopes in high temperature solutions sampled before and after ODP Leg 158 drilling at TAG hydrothermal field (26°N, MAR)

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**Abstract.** Hydrothermal fluids were sampled for dissolved gases at TAG (26°N - MAR) during two Alvin dive series, in April-May 1993 and March 1995, respectively 17 months before and 4 months after Ocean Drilling Program Leg 158. Total gas volumes extracted from the 1993 and 1995 samples are of the same order of magnitude, even if some increase in H<sub>2</sub>S, CO<sub>2</sub> and CH<sub>4</sub> was noticeable in the 1995 samples. No significant difference was observed in helium concentration and helium isotopic ratio (<sup>3</sup>He/<sup>4</sup>He=8.2 +/-0.1 Ra). The CH<sub>4</sub>/<sup>3</sup>He ratio found in black smokers sampled in 1993 is around  $9 \times 10^6$ , close to ratio found in MAR basalts. In 1995, this ratio is around 4 times higher, due to the CH<sub>4</sub> increase. The  $\delta^{13}\text{C}$  in CO<sub>2</sub>, measured on two black smokers sampled in 1993, is uniform at -8.4 to -8.8‰ (versus PDB), while a  $\delta^{13}\text{C}$  value of -13‰ is measured at a new site sampled in 1995, located 40 meters east of the main Black Smokers Complex (BSC). CH<sub>4</sub>/<sup>3</sup>He and <sup>13</sup>C values both point to a mainly magmatic (abiogenic) origin of the carbon species in the system. Overall comparison of the 1993 and 1995 data suggests that the hydrothermal circulation is continuing through the basaltic layer carrying helium, CO<sub>2</sub> and CH<sub>4</sub> of magmatic origin, even if some thermogenic CH<sub>4</sub> contribution seems likely in the 1995 samples collected in the new active area. We can speculate that the 1994 drilling may have modified the previously steady state hydrothermal circulation in the TAG mound and temporarily enhanced the hydrothermal circulation, by opening new pathways favouring transient input of CH<sub>4</sub> rich fluids, before coming back to a new steady state.

### Introduction and Background

The discovery of TAG hydrothermal field over the Mid-Atlantic Ridge in 1985 [Rona *et al.*, 1986] was the first direct evidence for high temperature venting on a slow spreading ridge. The first hydrothermal fluids collected in this area [Campbell *et al.*, 1988] showed compositions similar to those observed on the East Pacific Rise (EPR). High-temperature fluids sampled by Alvin expeditions in 1986, 1990 and 1993 [Campbell *et al.*, 1988; Edmond *et al.*, 1995], the BRAVEX cruise with the MIR

submersibles in 1994 [BRAVEX 94 scientific team, 1994], the MODE 94 cruise with the Shinkai 6500 [Fujioka *et al.*, 1994; Edmonds *et al.*, 1994; Gamo *et al.*, this issue], all point to the stability of temperature and chemical compositions although some geological modifications were perceptible, as indicated by the slowly decreasing activity at the Kremlin site. All these data were obtained before Leg 158 Ocean Drilling Program (ODP), conducted on the Joides Resolution (September 29 to November 22, 1994). The March 1995 diving cruise by R/V AtlantisII/Alvin was the first cruise conducted at TAG subsequent to drilling. One major objective was to study the influence of drilling on the fluid chemistry. Here, we report the analyses of gases and helium isotopes in TAG hydrothermal fluids collected during the 1993 and 1995 Alvin dives, respectively 17 months before and 4 months after drilling, compare them to other MAR hydrothermal vents and briefly discuss the influence of drilling on gas composition. In addition, data are reported for a limited number of  $\delta^{13}\text{C}$ CO<sub>2</sub> measurements to better characterize the origin of carbon in this hydrothermal system.

### Geological Setting

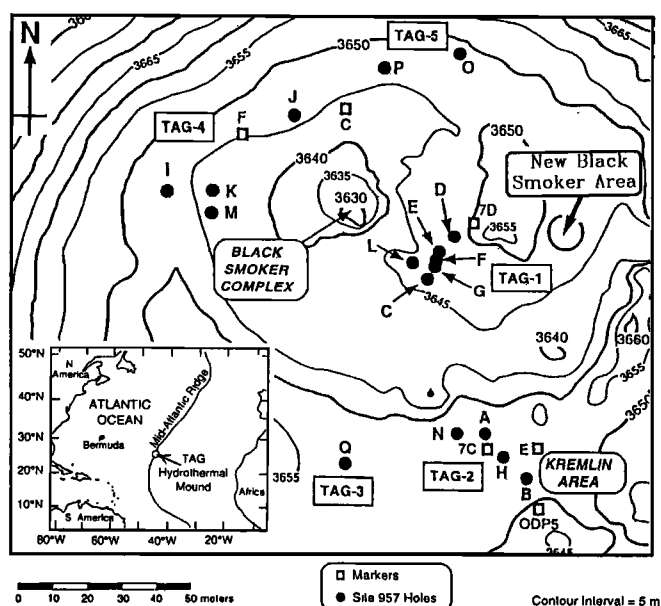
The TAG hydrothermal field occurs on the east wall and adjacent floor of the rift valley of the Mid-Atlantic Ridge near 26°N, 45°W. The target of ODP Leg 158 and of the work reported in this paper was a hydrothermally active mound about 200 m in diameter and 45 m high within the TAG field between depths of 3625 and 3670 m on the floor of the rift valley near the base of the east wall. During the ODP Leg 158, seventeen holes were drilled in five areas (TAG 1 to TAG 5) across the TAG hydrothermal field, including the active Black Smoker Complex (BSC) and the white smokers Kremlin area (Figure 1) [Humphris *et al.*, 1995].

### Methods

During the 1993 cruise, fifteen hydrothermal fluids sampled from BCS and Kremlin were collected for gas study. The samples were extracted on board immediately after collection by Alvin using a gas extractor ensuring total extraction of volatiles [Charlou *et al.*, 1993]. All gases were compressed into 100 ml gas sampling flasks with gas-tight valves, at a pressure above the atmospheric pressure to avoid air contamination. Laboratory tests show that more than 95% of the gases in solution are extracted

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**Figure 1.** General map of the TAG (Trans-Atlantic Geotraverse) hydrothermal field on the Mid-Atlantic Ridge and Seabeam bathymetry of the active TAG hydrothermal field showing the active TAG hydrothermal mound complex, the low-temperature portion of the active sulfide mound (Kremlin area) on the eastern rift-valley wall. The locations of the ODP Leg 158 drill holes at the active mound [TAG-1 to TAG-5] are shown. The newly active area where black smokers were sampled during the Alvin 1995 cruise is located 40 meters east of the active Black Smoker Complex [modified after *Humphris et al., 1995*].

from the fluid solution after 3 successive extraction/compression operations. In addition, a small aliquot of gas was sealed in a 1.8 ml volume Corning glass bulb for helium isotopes determination.

During the 1995 cruise, twenty high-temperature fluids, with a small number dedicated to gas analysis, were collected from two areas: the central Black Smoker Complex (BSC) and the newly active area of black smoker venting located 40 meters east of the BSC [*Scientific Party of Leg 132-02 of the Atlantis II, 1995*]. The fluids were transferred on board in two sets of 10 ml capacity copper tubes sealed with knife-edge clamps. The first set of tubes were extracted on shore using the IFREMER gas extractor. Due to the limited fluid volume, the gas was concentrated in 5 ml capacity metallic tubes for gas and carbon isotopes analysis. The second set of copper tubes containing the fluid were analysed at CEA-Saclay for helium isotopes.

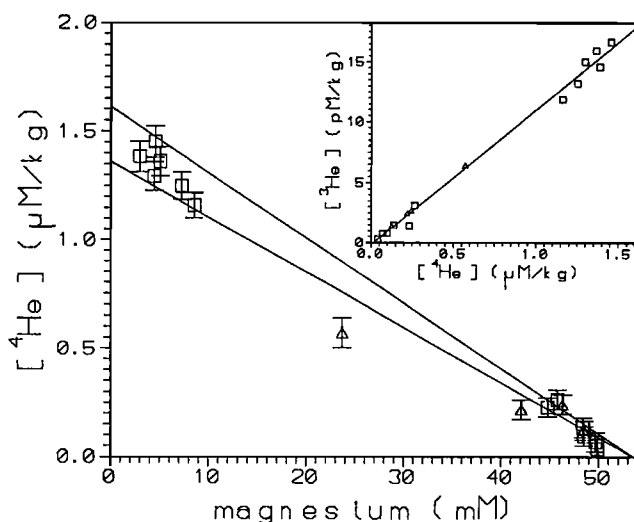
For both cruises, the helium analyses were carried out at CEA-Saclay with a VG-3000 mass spectrometer. Analytical precision on the helium isotope ratio measurement is better than  $\pm 1\%$ . Helium concentrations were determined from  $^4\text{He}$  peak intensity. Its overall uncertainty including both degassing efficiency and analytical uncertainty is estimated around  $\pm 5\%$ . Carbon isotopes analyses were also carried out at CEA-Saclay on a Mat-Finnigan 252 mass spectrometer coupled to a gas chromatograph.  $\delta^{13}\text{C}$  values, determined with a precision of  $\pm 0.1\%$  have been corrected for the contribution from ambient seawater. The gas measurements were carried out at IFREMER with a HP-5890 gas chromatograph equipped with a thermal conductivity detector associated with a flame ionisation detector on line. Mass balance of quantified gases matches the global pressure of the samples. Analytical precision for the different gases is between 1 to 3%.

## Results

As far as gas chemistry is concerned, a few data are available along the MAR hydrothermal sites, including TAG and Snakepit hydrothermal fields [*Charlou and Donval, 1993; Charlou et al., 1993; Rudnicki and Elderfield, 1992; Jean-Baptiste et al., 1991*], Lucky Strike ( $37^{\circ}17'N$ ) and Menez Gwen sites ( $37^{\circ}50'N$ ) [*Charlou et al., 1995*].

The helium concentrations in the 1993 samples show a linear trend with magnesium (Figure 2), thus indicating a rather constant helium endmember concentration for all the TAG vents. The  $^4\text{He}$  endmember value is in the range 1.30-1.60  $\mu\text{mol/kg}$  of fluid, that is  $\approx 800$  times the ambient seawater concentration.  $^3\text{He}$  versus  $^4\text{He}$  data are also shown in the inset (Figure 2). The slope of the correlation corresponds to a hydrothermal endmember  $^3\text{He}/^4\text{He}$  of  $8.2 \pm 0.1$  Ra. When compared to previous helium measurements by *Rudnicki and Elderfield (1992)*,  $^4\text{He} \approx 1.40$ - $1.64$   $\mu\text{mol/kg}$  of fluid and  $^3\text{He}/^4\text{He}$  ratio  $\approx 6.97$ - $8.52$  Ra, these results indicate no detectable variation over a 3 year period. These values are also close to those measured at the Snakepit (MAR,  $23^{\circ}N$ ) [*Jean-Baptiste et al., 1991*] indicating much similarity in the helium geochemistry between both sites.

Total gas volume (117 to 194 ml STP/kg) is found to be lower at TAG than at other hydrothermal active sites (Table 1). In the 1993 samples, major gases  $\text{H}_2\text{S}$  and  $\text{CO}_2$  represent 40% and 42% respectively of the total.  $\text{CO}_2$  (3.31 to 3.56 mmol/kg) and  $\text{H}_2\text{S}$  (3.2 to 3.8 mmol/kg) endmembers are close to the 1990 values. The 1993  $\text{CH}_4$  endmember (0.15 mmol/kg - 2% of total gas volume) is similar to the 1990 value, indicating stability of the hydrothermal system during the 1990-1993 period. The  $\text{CH}_4/^3\text{He}$  ratio, close to  $9.0 \times 10^{16}$ , is uniform in all fluids collected in 1993 from the BSC. This ratio is in the same order of magnitude as the ratio found in MAR basalts and supports the abiogenic origin of  $\text{CH}_4$  at TAG, as already observed in other hydrothermal sites [*Charlou and Donval, 1993*], and in agreement with the carbon isotope composition  $\delta^{13}\text{C}\text{CO}_2$  of two samples analysed in 1993,  $-8.8$  and  $-8.4 \pm 0.1\%$  respectively.



**Figure 2.**  $^4\text{He}$  versus magnesium and  $^3\text{He}$  versus  $^4\text{He}$  (inset). Open squares and triangles are for TAG 93 and TAG 95 fluids respectively. The slope of the correlation corresponds to a  $^3\text{He}/^4\text{He}$  average ratio of the hydrothermal endmember of  $8.2 \pm 0.1$  Ra. All samples, including those collected from black and white smokers during the 1993 and 1995 Alvin dives, are considered.

**Table 1.** Gas Endmembers in TAG 93 and 95 Black Smokers Compared to Fluids From Other Sites of the Mid-Atlantic Ridge

	ambient sw	Snakepit		TAG				Lucky Strike
		1988 (1)	1990 (2)	1990 (3)	1993 (this work)	1994 (4)	1995 (this work)	1993 (5)
T (°C), max	2.5	345	335-356	360-366	361	320	358	152-320
pH	7.8	3.8	3.7	3.35	3.30-3.38	3.0	3.4	3.5
Mg (mM)	53.2	0	0	0	0	0	0	0
Cl (mM)	549	559	563	636	649-665	675	643-645	450-500
Si (mM)	0.040	18.0	19.2	20.7	19.1-19.6	21.4	20.0	12.0-15.5
Tot. Gas (ml STP)	65	-	-	-	173-194	-	224*	300-750
H2 (mM)	0.004	-	-	-	0.18-0.23	-	-	0.02-0.07
CO2 (mM)	2.3	-	-	-	3.31-3.56	-	4.10	8.9-24.9
Ar (μM)	16	-	-	-	21.6-24.3	-	25	-
N2 (mM)	0.59	-	-	-	0.8-0.95	-	0.9	0.80-0.89
CH4 (mM)	0.5x10 <sup>-6</sup>	0.062	0.046-0.059	0.14-0.15	0.15-0.16	-	0.50-0.62	0.67-0.89
H2S (mM)	0	2.7	6.1	2.5-3.5	3.2-3.8	3.1	4.0	2.5-3.0
R/Ratm	1.00-1.07**	8.4	7.86+/-1.85	7.5+/-0.9	8.2+/-0.1	-	8.14	-
<sup>4</sup> He (μM)	1.8 x 10 <sup>-3</sup>	2.05	1.37	1.64	1.35-1.60	-	1.0	-
δ <sup>13</sup> C(CO2) (‰/PDB)	-	-	-	-	-8.4/-8.8	-	-6.9/-13.0	-
CH4/ <sup>3</sup> He	-	2.6 x 10 <sup>6</sup>	3.0-3.9 x 10 <sup>6</sup>	8.8 x 10 <sup>6</sup>	8.8 x 10 <sup>6</sup>	-	43-52 x 10 <sup>6</sup>	-

The SnakePit, TAG and Lucky Strike hydrothermal sites are located at 23°N, 26°N and 37°17'N respectively. For TAG 95 samples, Cl and Si data are from *H. Edmonds*. \* Total gas volume found in TAG 95 is corrected for H2 contribution. \*\*R/Ratm found in deep seawater near TAG [*Jenkins et al.*, 1980].  $R = {}^3\text{He}/{}^4\text{He}$  and  $\text{Ratm} = 1.38 \times 10^{-6}$ .

References: (1) *Jean Baptiste et al.*, 1991. (2) *Rudnicki and Elderfield*, 1992; *Edmond et al.*, 1995. (3) *Rudnicki and Elderfield*, 1992; *Edmond et al.* (1995). (4) *Gamo et al.*, this issue. (5) *Charlou et al.*, 1995.

Gas data in fluids sampled in 1995, after drilling, point to further modifications of the system. A reactivation of CH<sub>4</sub> degassing is observed. The CH<sub>4</sub> endmember in the 1995 fluids reaches 0.62 mmol/kg, 4 times the 1993 value.

Helium isotopic ratio is very comparable in the 1993 and 1995 samples (Table 1). This finding is not surprising since the helium isotopic ratio basically represents the signature of the magmatic source beneath the ridge and is not expected to vary on a short timescale in absence of a significant magmatic event. Helium concentrations are also close to previous measurements. However, since the 1995 samples were transferred from the titanium syringes directly into copper tubes on deck, one cannot exclude that the slight difference is due to some helium loss while sampling the fluid. The helium isotope data at TAG compare well with the values found at the Snakepit site (23°N) [*Jean-Baptiste et al.*, 1991; *Rudnicki and Elderfield*, 1992] and elsewhere at other known active hydrothermal sites, pointing to the great homogeneity of the helium composition of the upper mantle. After drilling, the CH<sub>4</sub>/<sup>3</sup>He ratio is 4 to 5 times higher (43 to 52 x 10<sup>6</sup>), suggesting a likely contribution of thermogenic methane. Some difference is also observed on the carbon isotope composition δ<sup>13</sup>C(CO<sub>2</sub>) especially at the new black smoker site, 40m east of the main TAG smokers, with a value of -13 ‰ for this very young site (Table 1).

## Discussion

### Pre-Drilling Evolution

Time series measurements of the composition of the TAG smokers between 1986 and 1990 show, as for Snakepit site, temporal stability [*Campbell et al.*, 1988; *Edmond et al.*, 1995]. The preliminary analyses of fluids collected in August 1994 with the submersible Shinkai 6500 prior to drilling by ODP do suggest

substantial changes in the white smoker fluid chemistry compared to samples taken in 1990, with, in particular, a marked increase in H<sub>2</sub>S concentrations (up to 5 mmol/kg), lower pH and alkalinity [*Edmonds et al.*, 1994; *Gamo et al.*, this issue]. An increased silica concentration was also observed in the area of hydrothermal diffuse flows [*Fujioka et al.*, 1994]. Steady state changes and accidental events were observed, with evidence of active micro seismicity during the observation period [*Fujioka*, 1995]. These chemical observations suggest that the circulation at the TAG hydrothermal field was evolving, with slight modifications in the plumbing system and water-rock reactions.

### TAG Drilling Effects

The drilling results from the TAG mound indicate a multi-stage depositional history, complex fluid interactions, and reveal the extent of subseafloor mineralization and alteration [*Herzig et al.*, 1995]. The high temperature upflow zone is not restricted to a single pipe-like feature but occurs as a complex network of channelways through a permeable sequence of pre-existing breccias. The chemistry of fluids sampled by extracting porewater from sediments collected from the periphery of the mound suggests that there are mixtures of high-temperature endmember fluids with ambient seawater that undergoes modification within the mound [*Mills and Tivey*, 1995]. Rapid temperature changes were observed during and immediately following the drilling program. Longer term changes in the mound hydrology are manifested by the increased activity to the E and NE of the mound coupled by a reduction in activity to the SW of the mound. One of the drillhole areas has developed a multitude of small Kremlin-type beehive chimneys which emit hot fluids (near 350°C) with chemistry indistinguishable from the BSC [*Scientific Party of Leg 132-02 of the Atlantis II*, 1995].

### Post-Drilling Fluid Chemistry

The temperature changes observed during and after drilling may have accelerated the liberation of gases (Table 1) through new pathways opened by the drilling. Increased CO<sub>2</sub> and CH<sub>4</sub> outgassing is generally clearly observed in areas under stress and subject to increase of the intensity of redox reactions and to fault movement caused by seismic earthquakes [Sugisaki *et al.*, 1983]. The gas variations, as well as the CH<sub>4</sub>/<sup>3</sup>He change, suggest some thermogenic production of CH<sub>4</sub> from organic matter. δ<sup>13</sup>C<sub>CO2</sub> values typical of the MORB reservoir are in the range -4 to -7.5 ‰ [Mattey *et al.*, 1984]. The lower value (-13 ‰) found at the new venting site, probably due to thermogenic decomposition of organic matter and bacterial oxidation of isotopically light organic matter (-25 ‰) to CO<sub>2</sub>, also points to an organic carbon component and supports the above interpretation. Permeability changes induced by new drilled pathways favour subsurface mixing between high temperature effluent and entrained surrounding seawater enriched in organic matter. The high temperature fluid may have decomposed hydrothermal animal tissue or microbial mats incorporated into the sulfide structure, generating gases of thermogenic origin released by the drilling operations. Submersible dives of May 1993 and August 1994, prior to drilling, showed that the internal plumbing system of the mound responsible for the venting of Kremlin fluids was progressively becoming sealed. This natural change within the system was modified by drilling operations, by initiating vigorous venting of primary hydrothermal fluids in the eastern part of the mound and influencing slightly the gas chemistry. Thus, we believe that drilling operations facilitated the release of CH<sub>4</sub> rich-fluids previously trapped in the shallow reaction zone and created new pathways permitting a temporary increase of the CH<sub>4</sub> outgassing. These additional flux of confined gases may be transiently released due to the disturbances resulting from the drilling operations.

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