Rare gases in mantle-derived rocks from Annobon Island: Preliminary step-heating helium results

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ABSTRACT

We report preliminary helium data obtained by step-heating and in-vacuo crushing of ultramafic xenolith whole rock and mineral separates collected from Annobon Island. For the first time, step-heating data reveals very large helium isotopic anomalies ($^3He/^4He$ up to $60R_a$, when normalized to the air ratio of 1.4×10^{-6}, $= 1R_a$) due to exposure of the xenoliths to cosmic rays. The olivine mineral separate records the highest cosmogenic ^3He (^3He_c) concentration ($(24.5 \pm 2.3) \times 10^{-18}$ mol/g). With a ^3He_c production rate of $(1.23 \pm 0.25) \times 10^{-22}$ mol/g/y, it would have taken an estimated 199,000 ± 19,000 years of surface exposure to produce the observed ^3He_c in the xenolith. This represents the first cosmonuclide age evidence for Upper Pleistocene volcanic activity in Annobon. Helium released by crushing is practically similar at 7.88 – $8.64R_a$ (mean = 8.28 ± 0.29 R_a) indicating that the Annobon magma source has a MORB-like $^3He/^4He$ ratio.

Key words: Annobon, xenolith, MORB-like He, cosmogenic ^3He, eruption age.

RESUME

Nous pressentons les données préliminaires sur hélium obtenir par 'step-heating' et 'in-vacuo crushing' des xenoliths ultramafigues (roche totale et minéraux), collectés à Annobon. Pour la premières fois, les données 'step-heating' montrent les larges anomalies en hélium ($^3He/^4He$ = $60R_a$, normaliser avec le rapport en air de 1.4×10^{-6}, $= 1R_a$), provenant de l'exposition des xenoliths aux rayons cosmiques. L'olivine enregistre la concentration de ^3He cosmogenique (^3He_c) le plus élevé ($(24.5 \pm 2.3) \times 10^{-18}$ mol/g). Avec un taut de production de ^3He_c de $(1.23 \pm 0.25) \times 10^{-22}$ mol/g/a, le temps d'exposition en surface pour produire le ^3He_c observer est estimé à 199,000 ± 19,000 ans, indiquant l'âge Pléistocène superoir d'activité volcanique à Annobon. L'hélium libéré par 'crushing' est pratiquement similaire à 7.88 – $8.64R_a$ (moyen = 8.28 ± 0.29 R_a), indiquant que la source magmatique d'Annobon est de gendre MORB.

Mots clés : Annobon, xenolith, hélium de MORB, ^3He cosmogenique, l'âge d'éruption.

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1. INTRODUCTION

Annobon is the most southwesterly of the Gulf of Guinea Islands that make up the oceanic part of the Cameroon Volcanic Line (CVL). The CVL is a prominent geologic feature in West-Central Africa that trends for about 1600 km from the Atlantic Ocean into mainland Africa (Fig. 1). Volcanic rocks of the CVL comprise of basalts, basanites, trachytes and phonolites, with a predominance of mafic rocks. A description of this volcanism can be found in Marzoli et al. (1999, 2000). The origin of the CVL is still a highly contentious topic among earth scientists (see recent debates in Halliday et al., 1990; Barfod et al., 1999; Aka et al., 2004). Rare gas studies can help to elucidate the dynamics of CVL mantle because important diagnostic variations exist in terrestrial ^{3}He/^{4}He isotopic ratios that allow an unmistaken distinction to be made between crustal, MORB, high-μ (HIMU) and 'high ^{3}He/^{4}He hotspot' reservoirs. We are currently carrying out a study on the rare gas inventory of the CVL mantle (Aka, 2000; Aka et al., 2001). This has implications on its origin. In this paper, we give a preliminary report of the results of step heating extractions of helium from mantle xenoliths and phenocrystic phases in basalts from Annobon. These are compared to helium contained in fluids that are released from the same phases by crushing (Aka et al., 2004). It is shown that the helium in these samples contains both a MORB-like and surface-derived cosmogenic components.

2. GEOLOGY OF ANNOBON ISLAND AND SAMPLING

Annobon (Fig. 2) is the smallest of the CVL islands. Its geology is described by Cornen and Maury (1980). The volcanic succession is made up of hyaloclastite breccias thought to represent the island-forming submarine eruptive phase. Basaltic flows cover most of the island. Lava from this unit has yielded an Ar/Ar age of 4.8 Ma (Lee et al., 1994). The lava pile is intruded by tristanite and trachytic plugs and basanitic dikes, K-Ar-dated at 5.3 - 3.9 Ma (Cornen and Maury, 1980). Around Annobon town north of the Island, pyroclastic material underlies a peridotite-bearing lava flow. The peridotite, sampled 50 m above sea level at 1°24'06"S, 5°37'52"E, is a composite xenolith with spinel lherzolite as the main xenolithic phase. It contains orthopyroxenite layers with a cumulate-like structure. The host rock is a basanite and was K-Ar-dated at 0.20 ± 0.01 Ma (Aka et al., 2004). Helium was extracted from olivine separates from the lherzolite

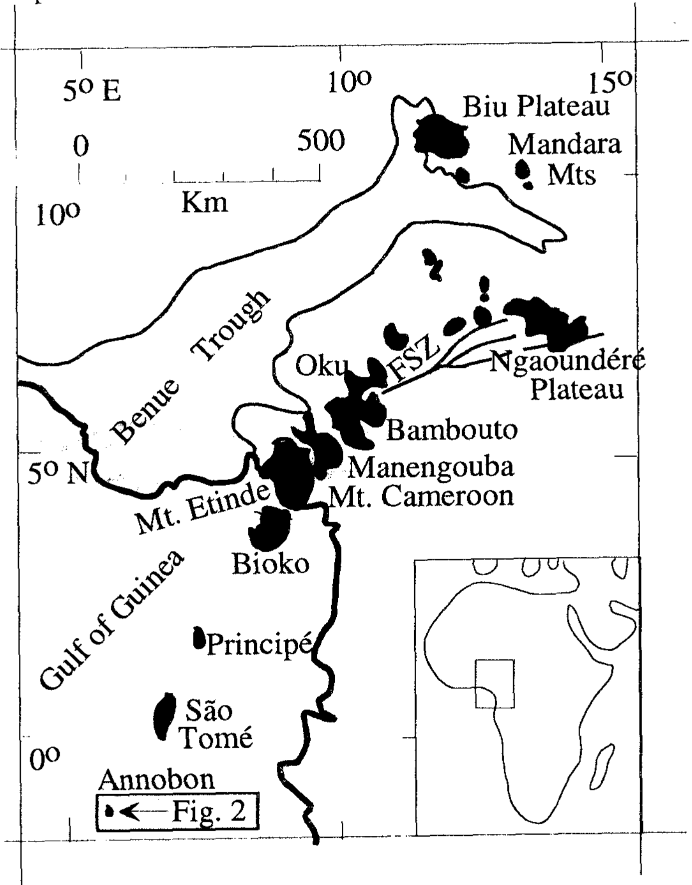

Figure 1: Map of the Cameroon Volcanic Line in the Gulf of Guinea. Main Cenozoic volcanic centers are indicated in black. Detail map of Annobon Island is shown in Fig. 2. FSZ stands for Foumban shear zone.

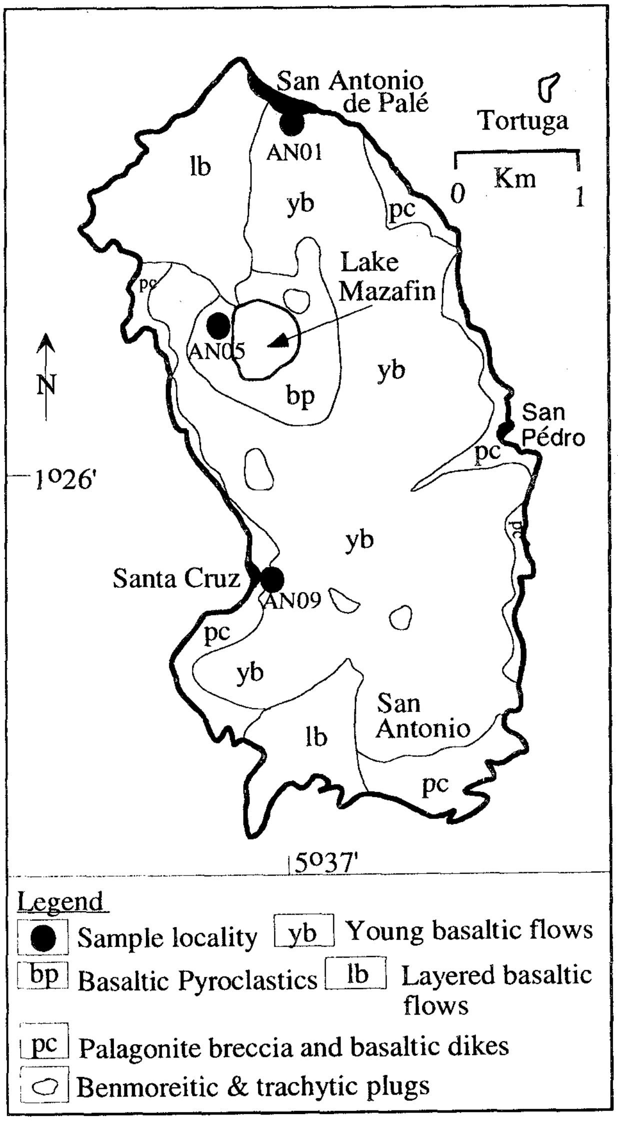

Figure 2: Geological map of Annobon Island (modified from Cornen and Maury, 1980) showing the location of samples.

(12AN01) and the orthopyroxenite whole rock (16AN01). A trachy-basalt collected near Santa Cruz was also K-Ar-dated and gave an age of 0.08 ± 0.01 Ma. K-Ar ages of 0.2 - 0.08 Ma in Annobon suggest that the last phase of volcanic activity in the island is < 1 Ma. However, what still remains an open question is when volcanic activity initially started (age of initiation) in the island.

3. EXPERIMENTAL METHODS

Xenoliths were hand-crumbled or lightly crushed in a steel mortar while lava chunks were crushed to 5 – 10 cm chips before further jaw-crushing to 0.15 - 1 mm. Minerals were separated by handpicking and magnetic separation techniques. Helium was extracted by step heating and in vacuo crushing. In step-heating experiments, sample was loaded into the sample holder and baked at 150 – 200 °C overnight for 24 hours to remove adhering atmospheric gases. Helium was extracted by step heating over a temperature range of 600 – 1800 °C and also by crushing. Details of crushing extractions are described in Aka et al. (2004). Helium was measured using MS-III, a magnetic sector type modified VG5400 mass spectrometer at the Laboratory for Earthquake Chemistry, University of Tokyo. ^{3}He and ^{4}He ion beams were detected on a double collector system, ^{3}He by electron multiplier used in ion-counting mode and ^{4}He by the high Faraday collector (feedback resistor = 10 GΩ). A resolving power of 600 allowed the complete separation of $^{3}He^{+}$ beam from H_3^{+} and HD^{+} beams. Measured ^{3}He/^{4}He ratios were calibrated with a standard ^{3}He/^{4}He gas (^{3}He/^{4}He = 1.71 x 10^{-4}). Furnace blanks for ^{4}He for the 600 – 1800 °C ranged from (9 – 35) x10^{-10} atoms. Crusher blanks (using same procedure as samples) for ^{4}He ranged from (8.7 – 18.3) x 10^{-10}-cm^{3}STP and for ^{3}He were <5 x 10^{-15} cm^{3}STP. Measured helium abundances have an uncertainty of 5% based on repeated analyses of the air standard and stability of the sensitivity of the mass spectrometer. Errors on the helium isotopic measurements are based on the statistical variation of the counting rate of ^{3}He detection.

4. RESULTS AND INTERPRETATION

4.1. Helium abundances and isotopic ratios

Results are displayed in Table 1 and plotted on Fig. 3. Salient features of the data are that (1) Duplicate analyses of the whole rock orthpyroxenite gave reproducible results of 34 and 37 (x10^{-13} mol/g) ^{4}He but the ^{4}He in the olivine separate is ~6 times lower. This is reflected in lower (10.7R$_a$) ^{3}He/^{4}He isotopic ratio in the pyroxenite than in the olivine (36.3R$_a$), indicating a marked ^{3}He/^{4}He isotopic disequilibrium between the pyroxenite and the olivine. (2) ^{3}He/^{4}He ratios for step heating experiments range from 8.7R$_a$ to ~60R$_a$. The highest measured ratio is ~2 times higher than the 37R$_a$ that has yet been observed in the most primitive terrestrial reservoir (Hilton et al., 1999). (3) In-vacuo crushing of the phases allowed extraction of gases with ^{3}He/^{4}He ratios that are practically similar at 7.88 – 8.64R$_a$ (see Fig. 3 and Aka et al., 2004), again showing isotopic disequilibrium between step heating and crushing results. Note that crushing rocks of different lithologies yielded ^{3}He/^{4}He ratios which are indistinguishable from each other within analytical uncertainty (Table 1 and Fig. 3). This is a strong indication that the source below Annobon and the oceanic lithosphere from which the

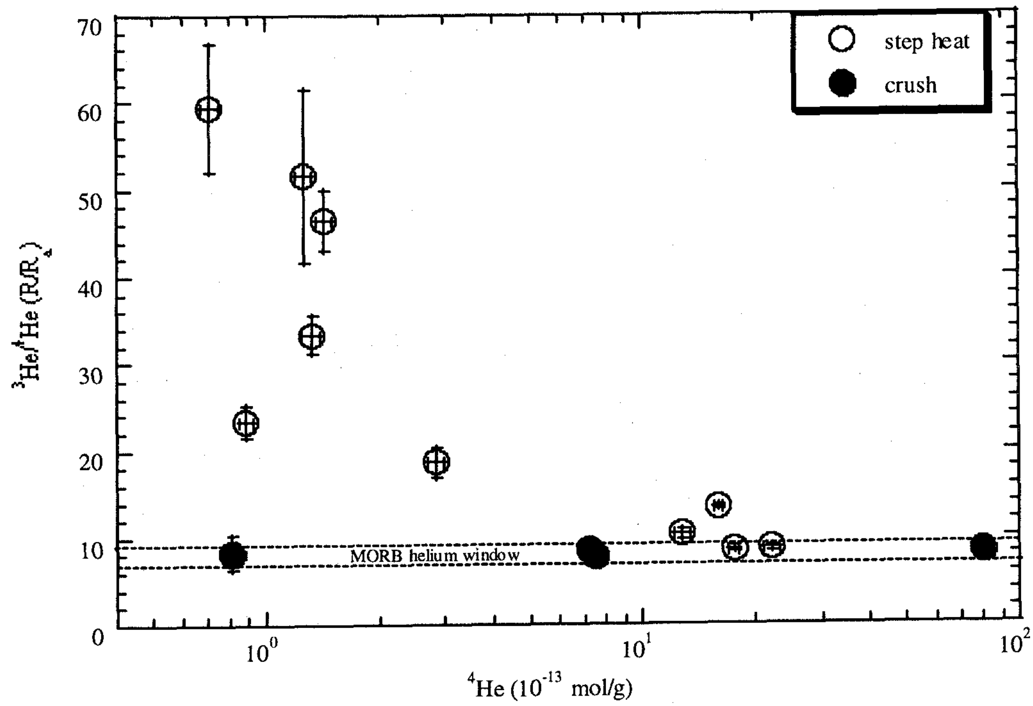

Figure 3: Comparison of step heating (this work) and crushed-released (Aka et al., 2004) ^4He concentrations against $^3He/^4He$ ratios for mantle-derived rocks from Annobon Island. Note that while step heating releases helium with R/R_a that ranges from MORB-like (8 ± 1R_a) to very high values, indicating a mixture of MORB and cosmogenic helium, crushing releases helium that plots within the MORB window.

xenoliths were picked up has similar (trapped) $^3He/^4He$ ratios. From the observed isotopic homogeneity in (crushed) $^3He/^4He$ ratios, it is suggested that a magma source with a unique $^3He/^4He$ ratio is of ~8.28 ± 0.29 R_a (1σ) is supplying Annobon.

4.2 Why is the $^3He/^4He$ ratio in the orthopyroxenite lower than in the olivine?

The $^3He/^4He$ isotopic disequilibrium between the pyroxenite and the olivine may be due to crustal (magma chamber) contamination by radiogenic ^4He or to post-eruptive radiogenic ^4He in-growth. Since the xenoliths carried to the surface by alkali basalts arrived unresorbed, their transport times must necessarily be short, less than 100h (e.g., Scarfe and Brearly, 1987). This suggests that they did not spend enough time in crustal magma chambers for radiogenic ^4He to grow.

An evaluation of the second mechanism requires knowledge of the U and Th concentrations in these phases which were not measured. Aumento and Hyndman (1971) measured U in primary orthopyroxenes and olivines separated from ultramafic xenoliths of oceanic upper mantle origin. If it is assumed that their reported values of 1 and 0.03 ppm U in the orthopyroxene and olivine respectively apply in this case, the production formula of Craig and

Table 1Step-heating released helium concentrations and isotopic ratios in mantle-derived rocks from Annobon Island

Lithology	Phase	Sample ID	Locality	Wt (g)	Temp (°C)	^4He 10^{-13} mol/g	±	^3He 10^{-18} mol/g	±	$^3He/^4He$ (R/Ra)	±	^3He_c [§] 10^{-18} mol/g	±
xenolith	wr orthopyroxenite	16AN01B	San Antonio	2.178	600	0.89	0.04	2.93	0.27	23.45	1.85	1.89	0.28
					900	1.33	0.07	6.23	0.51	33.40	2.19	4.68	0.52
					1200	12.77	0.64	18.42	1.50	10.31	0.66	3.57	1.77
					1500	21.97	1.10	26.88	1.68	8.74	0.33	1.31	2.34
					1800	bd							
					Total	36.96	1.27	54.46	2.33	10.52	0.58	11.45	3.24
Replicate				2.108	1200	15.96	0.48	29.99	1.05	13.42	0.24	11.42	1.40
					1800	17.54	0.53	21.28	0.97	8.67	0.30	0.88	1.40
					Total	33.50	0.71	51.27	1.43	10.93	0.38	12.30	2.26
xenolith	olivine	12AN01	San Antonio	2.049	900	0.71	0.04	5.88	0.79	59.34	7.35	5.06	0.79
					1200	1.43	0.07	9.27	0.83	46.42	3.43	7.61	0.83
					1500	1.27	0.06	9.17	1.82	51.48	9.90	7.69	1.82
					1800	2.85	0.14	7.46	0.77	18.66	1.70	4.13	0.80
					Total	6.26	0.18	31.78	2.28	36.25	2.80	24.50	2.31

[§] = cosmogenic ^3He. bd = below detection. wr = whole rock.

Lupton (1976) can be employed to make a rough estimate of the helium produced in the xenolith after eruption:

$J_{(He)} = (0.2355 \times 10^{-12} \times U\ (ppm) \times ((1 + 0.123\ (Th/U - 4))$.

For a Th/U ratio of 3.3, this gives a production rate of 9.6×10^{-18} mol/g/y. In 0.2 Ma, 1.9×10^{-12} mol/g ^4He could be produced in the rock. For olivine, a production rate of 2.9×10^{-19} mol/g/y is obtained, giving a ^4He of 5.8×10^{-14} mol/g. While the estimated ^4He is similar to the observed value in the orthopyroxenite (Table 1), it is an order of magnitude lower in the olivine. This suggests that radiogenic ^4He in-growth may have perturbed the helium of the pyroxenite.

4.3 MORB-like helium in Annobon mantle source

Helium isotopic disequilibrium between step heating and crushing suggests the release of fluids from different sites, the high $^3He/^4He$ ratios corresponding to matrix hosted helium and the low $^3He/^4He$ ratios to melt/fluid inclusion-hosted helium. Elemental and isotopic release systematics for the xenolithic olivine (Fig. 4) which indicate decreasing $^3He/^4He$ ratios with increasing temperature support this view. The estimated magmatic $^3He/^4He$ ratio for Annobon overlaps the range observed in N-MORB that has a $^3He/^4He$ ratio of $8 \pm 1R_a$ (Graham et al., 1992). This suggests that Annobon is supplied with an asthenospheric (MORB) helium having a constant $^3He/^4He$ ratio. Published Sr, Nd and Pb isotopic data (summarized in Aka, 2000) for basanites and *hy*- normative basalts from Annobon range from $0.70325 < ^{87}Sr/^{86}Sr < 0.70374$; $0.512858 < ^{143}Nd/^{144}Nd < 0.512972$ and $18.868 < ^{206}Pb/^{204}Pb < 19.272$ respectively. These values are more radiogenic or less depleted (Sr and Pb) than typical Atlantic N-MORB (see Rollinson, 1993). This raises a problem that the N-MORB-type helium isotopic ratios in Annobon appear to be linked with rather more enriched lithophile isotopic ratios, suggesting a decoupling of helium from the lithophile elements. To address this issue appropriately, lithophile element isotopic ratios will have to be measured in the same phases like the helium isotopes. Work is underway in this direction.

4.4 What is the origin of high $^3He/^4He$ ratios in the xenoliths?

Pioneering studies by Kurz (1986) showed that two dominant mechanisms can generate high $^3He/^4He$ isotopic ratios in terrestrial samples: spallation reactions involving cosmic ray secondary particles during exposure of the sample on the earth's surface and neutron capture through the reaction $^6Li(n,\alpha)^3H \rightarrow {}^3He$. The first mechanism is considered to be responsible for the elevated $^3He/^4He$ ratios in Annobon xenoliths for the following reasons. (1) Of the elements with the largest neutron capture cross sections, thus greatest contributors to ^3He via the second mechanism, Li, B, Mg and Si are the most significant in ultramafic rocks. Assuming a pyrolite earth model composition for Li and B of 1.5 and 0.3 ppm respectively (McDonough and Sun, 1995), and using the whole rock chemical composition (wt. %) of sample AN01 (Aka, 2000) of $SiO_2 = 52.7$, $TiO_2 = 0.46$, $Al_2O_3 = 4.49$, FeO = 11.2, MnO = 0.18, MgO

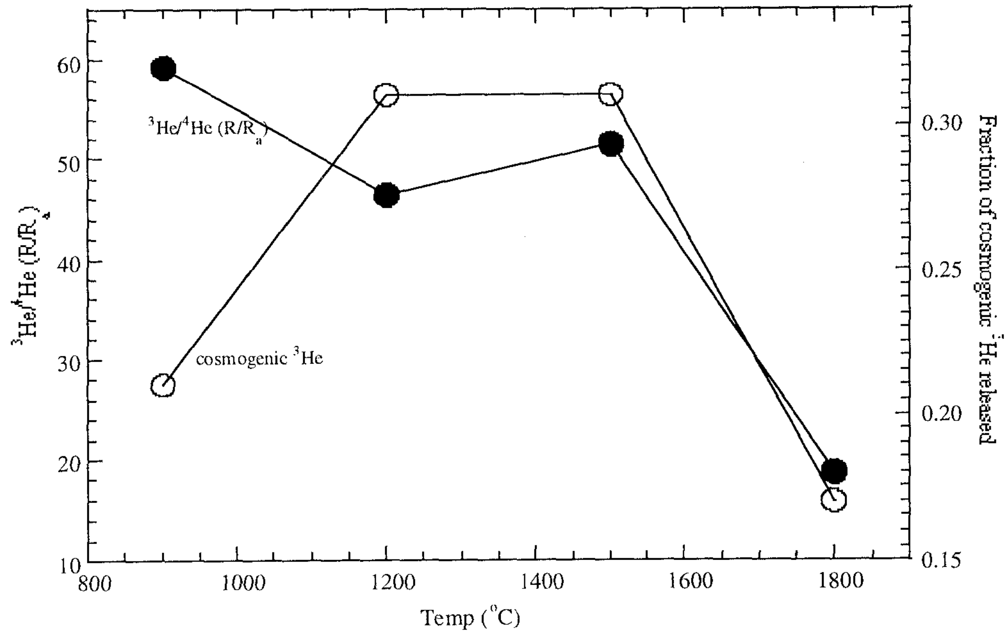

Figure 4: Temperature-dependent release sytematics of helium in xenolithic olivine from Annobon Island.

= 27.04, CaO = 3.35, Na_2O = 0.35, K_2O = 0.05 and P_2O_5 = 0.04, the contribution by this mechanism is evaluated for this sample using equation 7.4 in Mamyrin and Tolstikhin (1984), and neutron capture cross-section data from their Table 7.1. The result shows a calculated $^3He/^4He$ ratio of only 1.6×10^{-9} or $1.1 \times 10^{-3} R_a$, giving a measured to calculated $^3He/^4He$ ratio in the sample of $\sim 10^4$. It is concluded that nucleogenic generation of ^3He plays no role in the high $^3He/^4He$ ratios observed in the xenoliths. (2) The helium isotopic composition of fluids released by crushing (Aka et al., 2004) is an order of magnitude lower than that of fluids released by heating the same phases. In-vacuo crushing released fluids with helium isotopic composition an order of magnitute lower than in heating. This shows that crushing released helium from fluid/melt inclusions while heating released matrix-hosted helium. The latter has a cosmogenic origin (Kurz, 1986).

Cosmogenic ^3He (^3He_c) is calculated from:

$^3He_c = {}^3He_m - {}^3He_i$, where

$^3He_i = {}^4He_m \times ({}^3He/{}^4He)_i$

with 'm' = measured and 'i' = initial He trapped in fluid inclusions and released by crushing. A $({}^3He/{}^4He)i$ value of $8.31 \pm 0.33 R_a$ (Aka et al., 2004) which is similar to the ratio estimated for Annobon source above, was used for the calculation. ^3He_c calculated in this way ranges from $(\sim 1 - 8) \times 10^{-18}$ mol/g, and constitutes up to 86% of the total ^3He measured in the xenoliths (Table 1). Note that the ^3He_c in the olivine separate is 2 times more than in the orthopyroxenite whole rock. Because helium abundances in the orthopyroxenite might have been perturbed by radiogenic ^4He, this sample will not be considered further in the discussion.

4.5 Cosmogenic ^{3}He and xenolith eruption age

The production rate of a cosmonuclide in olivine can be calculated at a particular location if the eruption age of the sample is constrained. In this way, the cosmonuclide can be used as a geochronometer (Kurz, 1986). Dunai and Wijbrans (2000) have published a cosmogenic ^3He production rate of $(2.0 \pm 0.1) \times 10^{-22}$ mol/g/yr at sea level and high latitudes. We scale this value by a factor of 0.617 to the Annobon xenolith altitude (1027 g/cm^2 atmospheric depth) and geographic latitude (see paragraph 2) using the polynomial fits of Lal (1991). This gives a ^3He_c production rate of $(1.23 \pm 0.25) \times 10^{-22}$ mol/g/y (1σ uncertainty of ±20%). At this rate, it would have taken 199,000 ± 19,000 years to produce the observed ^3He_c in the xenolith olivine (Table 1). This represents the ^3He_c eruption age of the xenolith, assuming that since eruption, the xenolith has not been affected by uplift or subsidence, has suffered no erosion and has not lost or gained helium except by in situ cosmogenic production. These assumptions seem justified if the calculated ^3He_c age is compared with the 200,000 ± 10,000 year K-Ar age of the xenolith host basanite (Aka et al., 2004). The 2 ages are similar within analytical error and constitute the first cosmogenic age evidence for Upper Pleistocene volcanic activity in this island.

5. CONCLUSION

Preliminary helium results of step heating and extraction of rare gases from ultramafic whole rock and mineral separates are reported for Annobon Island and compared to results obtained by crushing the same phases. Step heating data show very high $^3He/^4He$ ratios, up to $60R_a$ in individual temperature steps of the olivine separate, while crushing data shows that the island is supplied with helium having an asthenospheric (MORB-like) signature. The high $^3He/^4He$ ratios result from exposure of the xenoliths to cosmic rays. The observed cosmogenic ^3He abundance indicates that the xenoliths may have been exposed to cosmic rays since Upper Pleistocene time.

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