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Research Article

^{3}He/^{4}He Ratio in Olivines from Linosa, Ustica, and Pantelleria Islands (Southern Italy)

Elise Fourré,[1] Patrick Allard,[2] Philippe Jean-Baptiste,[1]
Dario Cellura,[3] and Francesco Parello[3]

[1] LSCE, CEA-CNRS-UVSQ, Centre de Saclay, 91191 Gif-sur-Yvette, France
[2] IPGP, CNRS, 75005 Paris, France
[3] CFTA, Università di Palermo, 90123 Palermo, Italy

Correspondence should be addressed to Philippe Jean-Baptiste, philippe.jean-baptiste@cea.fr

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We report helium isotope data for 0.03–1 Ma olivine-bearing basaltic hawaiites from three volcanoes of the southern Italy magmatic province (Ustica, Pantelleria, and Linosa Islands). Homogenous ^{3}He/^{4}He ratios (range: 7.3–7.6 R_a) for the three islands, and their similarity with the ratio of modern volcanic gases on Pantelleria, indicate a common magmatic end-member. In particular, Ustica (7.6 ± 0.2 R_a) clearly differs from the nearby Aeolian Islands Arc volcanism, despite its location on the Tyrrhenian side of the plate boundary. Although limited in size, our data set complements the large existing database for helium isotope in southern Italy and adds further constraints upon the spatial extent of intraplate alkaline volcanism in southern Mediterranea. As already discussed by others, the He-Pb isotopic signature of this magmatic province indicates a derivation from a mantle diapir of a OIB-type that is partially diluted by the depleted upper mantle (MORB mantle) at its periphery.

1. Introduction

Plio-Quaternary volcanism in the Italian Peninsula has developed in the complex tectonic environment of African-European continental plate collision and Adriatic-Ionian slab subduction under the expanding Tyrrhenian Sea back-arc basin ([1] and references therein). This complexity is reflected in the wide compositional diversity of erupted magmas, which range from ultrapotassic and potassium-rich in Tuscany and the Roman-Napolitean province, to calc-alkaline in the Aeolian Island arc, and to OIB-type Na-alkaline basaltic in southern Sardinia and Sicily. This magmatic diversity is also accompanied by spectacular regional gradations in the trace-element and isotope geochemistry of volcanic products. In particular, the progressive south to north trend of ^{87}Sr/^{86}Sr and ^{18}O/^{16}O in magmas, and ^{13}C/^{12}C and ^{3}He/^{4}He in emitted gases [2–6] indicates a northward increase in contamination of magma sources by crustal material from the subducting Adriatic and Ionian plates.

Na-alkaline mafic magmatism currently active at or close to the collision plate boundary in southern Italy displays minor imprint of these subduction processes and thus provides the "cleanest" signature of the mantle beneath the region. This basaltic magmatism has developed on tensional tectonic faults cutting the African plate margin and over both a thinner crust and lithosphere (60–100 km). This basalt displays trace-element patterns typical of Ocean Island Basalts (OIB), relatively radiogenic ^{87}Sr/^{86}Sr and ^{206}Pb/^{204}Pb isotopic signatures [7–11], and has been recognized to represent a mantle component common to all Plio-Quaternary volcanic series in Mediterranea and western-central Europe [12–14]. However, the genesis of this OIB-type magmatism in a context of continental plate collision remains an enigma, for which have been proposed various, often contradictory, interpretative models (e.g., [15–17]; see [18] for a review).

The ^{3}He/^{4}He ratio is a well-known powerful tracer of the origin of magmas and of possible crustal contamination during magma storage and ascent (e.g., [19, 20]). ^{3}He (essentially primordial) and ^{4}He (produced by the

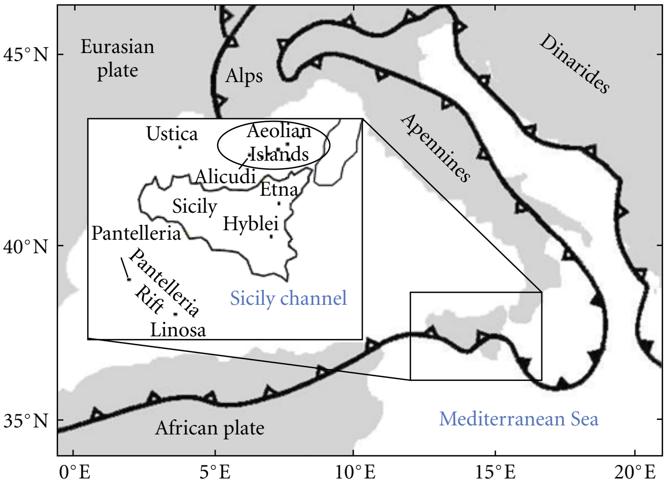


Figure 1: Location map of the study area.

Table 1: Sample description (ages for Linosa, Pantelleria, and Ustica are from Lanzafame et al. [38], Civetta et al. [39], and De Vita et al. [36], resp.).

Site	LINOSA			PANTELLERIA	USTICA	
Sample	Lin 15	Lin 20	Lin 27	Pant 8A	Ust 1	Ust 2
Latitude (N)	35°52′10.8″	35°52′11.3″	35°52′29.7″	36°48′53.3″	38°42′37.0″	38°42′42.5″
Longitude (E)	12°52′20.3″	12°52′01.5″	12°52′21.3″	11°55′37.8″	13°10′16.6″	13°10′06.6″
Rock type	Massive lava	Vesiculated lava	Vesiculated lava	Vesiculated lava	Coarse grain tuff	Fine grain tuff
Sampling site	Montagna Rossa	Montagna Rossa	Montagna Rossa	Cala dell'Alca	Monte Costa del Fallo	Monte Costa del Fallo
Type of outcrop	Lava flow	Lava flow	Lava flow	Lava flow	Pyroclastic flow	Pyroclastic flow
Age (Ma)	0.53–1.06	0.53–1.06	0.53–1.06	0.08–0.1	0.42–0.52	0.42–0.52

radioactive decay of U and Th) have distinct origins and varying proportions in Earth's reservoirs. Typical ^{3}He/^{4}He ratios vary from <0.1 R_a in continental crust (R_a is the atmospheric ratio equal to 1.38×10^{-6}), to 8 ± 1 R_a on average in the upper mantle, and up to ~40–50 R_a in products of plume-related ocean islands, such as Hawaii and Iceland [21, 22]. Moreover, various observations, such as He isotopic similarity in solid and gas phases at single volcanoes (e.g., [5, 23]), indicate no or minor ^{3}He/^{4}He fractionation during the physical processes of magma genesis, differentiation, and degassing and, therefore, can reliably be used to track magma sources.

Here, we report new results for He isotopes in olivine-bearing basaltic lavas from Ustica and Pantelleria that complement previously published data for these two islands, and ^{3}He/^{4}He results for Linosa, for which no data is currently available. This new data set complements a substantial existing database for helium isotopes in southern Italy and provides additional information about the spatial extent of intra-plate alkaline volcanism in southern Mediterranea.

2. Samples and Analytical Procedures

Ustica is located to the northwest of Sicily (Figure 1), and is made up of both subalkaline and alkaline basalts erupted between 750 and 130 ka [1]. Linosa and Pantelleria are located along the northwest to southeast rift system of the Sicily Channel, which separates the Sicilian platform from Africa (Figure 1). Most Linosa volcanic lithologies (1.06–0.53 Ma; [35]) are Na-alkaline to slightly transitional, mainly represented by basalts and hawaiites. The Pantelleria basalts exhibit a more complex volcanic history, which has produced mafic magmas (300–5 ka; [1]) from weakly alkaline to transitional, dominated by more evolved compositions such as trachytes, rhyolites, and pantellerites.

We analysed 0.5 to 2 mm olivine phenocrysts contained in unaltered samples of 0.03 to ~1 Ma old pyroclastites and basaltic lava flows, some with hawaiitic composition, from the three volcanic islands [11, 36]. Sample description is given in Table 1. Olivine crystals were handpicked under a binocular microscope from the crushed whole rocks, and altered grains or those with adhering glass were carefully removed. The pure olivine separates were precleaned with distilled water and acetone in an ultrasonic bath and dried for 15 minutes at 70°C under vacuum.

The weighed dry crystal samples (500 mg) were then loaded in a pneumatically actuated all-metal crusher. The crusher was connected to the inlet line of a MAP 215-50 noble gas mass spectrometer (CEA-Saclay facility; [37]), and pumped to ultra high vacuum overnight. After blank measurement, helium was extracted by stepwise crushing, each crushing cycle consisting of 100 strokes. Crushing cycles

Table 2: Measured He concentrations and isotope ratios in olivine crystals from basaltic-hawaiitic lava flows from Linosa, Pantelleria and Ustica volcanic islands.

Sample	Weight (g)	Number of strokes cumulative	R/R_a	^{4}He cumulative (10^{-8} cm^3 STP/g)	R/R_a cumulative
Linosa					
Lin 15	0.50214	100	7.27	3.58	7.27
		200	7.82	**3.96 ± 0.04**	**7.3 ± 0.2**
Lin 20-1	0.51271	100	7.53	4.23	7.53
		200	7.36	5.06	7.50
		300	6.43	5.40	7.44
		400	7.52	**5.61 ± 0.04**	**7.4 ± 0.2**
Lin 20-2	0.49958	100	7.57	4.04	7.57
		200	7.55	4.91	7.56
		300	7.53	5.38	7.56
		400	7.54	**5.62 ± 0.04**	**7.6 ± 0.2**
Lin 27	0.50058	100	6.99	4.48	6.99
		200	7.26	4.77	7.00
		300	7.01	**4.90 ± 0.04**	**7.0 ± 0.2**
Pantelleria					
Pant 8A	0.54177	100	7.65	0.249 ± 0.005	7.6 ± 0.3
Ustica					
Ust 1	0.50126	100	7.68	11.49	7.68
		200	7.56	13.00	7.67
		300	7.80	13.96	7.68
		400	7.21	14.61	7.66
		500	8.14	15.05	7.67
		600	7.97	15.40	7.68
		700	8.24	**15.7 ± 0.1**	**7.7 ± 0.2**
Ust 2	0.50146	100	7.67	14.85	7.67
		200	7.43	17.34	7.63
		300	7.61	18.43	7.63
		400	6.96	19.31	7.60
		500	7.40	19.73	7.60
		600	7.60	20.18	7.60
		700	7.58	20.65	7.60
		800	7.54	**20.9 ± 0.1**	**7.6 ± 0.2**

were renewed until the amount of extracted gas was close to the procedural blank ($2.5 \pm 1.5 \times 10^{-10}$ cm^{3}STP ^{4}He). In all samples, the neon content was at the blank level ($< 2 \times 10^{-10}$ cc STP), thus indicating no air contamination. ^{3}He/^{4}He ratios were determined in reference to a routinely used air standard performed before and after each sample. The reproducibility on the air standard was better than 0.4%. Instrumental uncertainties for ^{3}He/^{4}He ratio and helium concentration were in the range of 0.5% to 2% and 3% to 4% respectively, depending on the amount of gas available for mass spectrometry (see Table 2).

3. Results

The total helium concentrations in our sample set vary by two orders of magnitude (Table 2), from 2.49×10^{-9} cm^3 STP g^{-1} (Pantelleria) to 2.09×10^{-7} cm^3 STP g^{-1} (Ustica). Samples from Linosa display an intermediate, very homogenous He content averaging $4.8 \pm 0.8 \times 10^{-8}$ cm^3 STP g^{-1}. Analysis of duplicate samples Lin 20-1 and Lin 20-2 yielded identical concentration (5.6×10^{-8} cm^3 STP ^{4}He g^{-1}), within experimental uncertainties. The observed range in He content actually correlates with the differences in bulk crystal sizes. In particular, all olivines in Ustica samples were ≥1 mm (30% >1.4 mm), whereas 86% of olivines in Pantelleria sample were <1 mm.

Figure 2 shows that ~80% of the recovered helium was released during the first and second crushing steps, indicating that most of the entrapped helium resided in fluid inclusions or/and at grain boundaries of the olivine crystals. The fairly homogeneous ^{3}He/^{4}He ratio of residual helium recovered during subsequent crushing cycles (Figure 2) indicates no or minor release of either radiogenic or cosmogenic He from the crystal lattice. This is confirmed by the fact that the 0.03 Ma old Pantelleria olivines, albeit having the lowest He concentration, yielded a ^{3}He/^{4}He value similar to modern

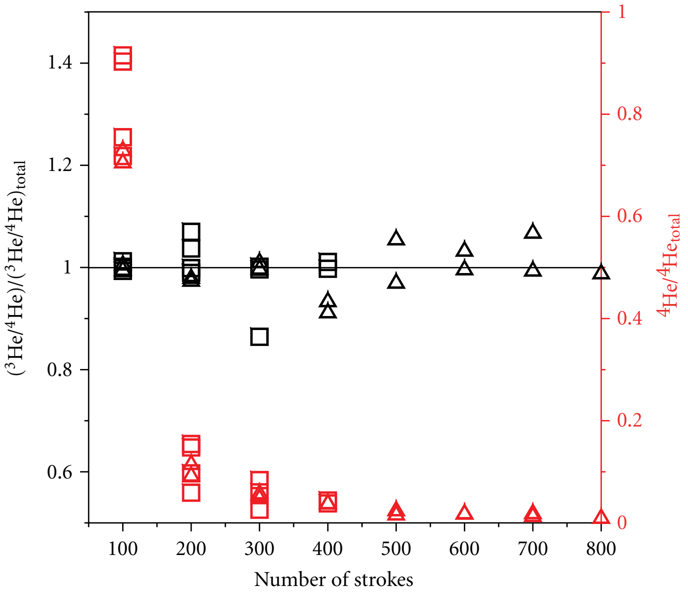


FIGURE 2: Fraction of the total helium released and $^3He/^4He$ trend as a function of the number of strokes for Linosa (squares) and Ustica (triangles) samples (N.B. due to its low He content, Pantelleria sample had only one stroke cycle).

volcanic gases [24]. Therefore, we can safely conclude that the measured $^3He/^4He$ ratios characterize essentially pure magmatic helium trapped within the olivine crystals.

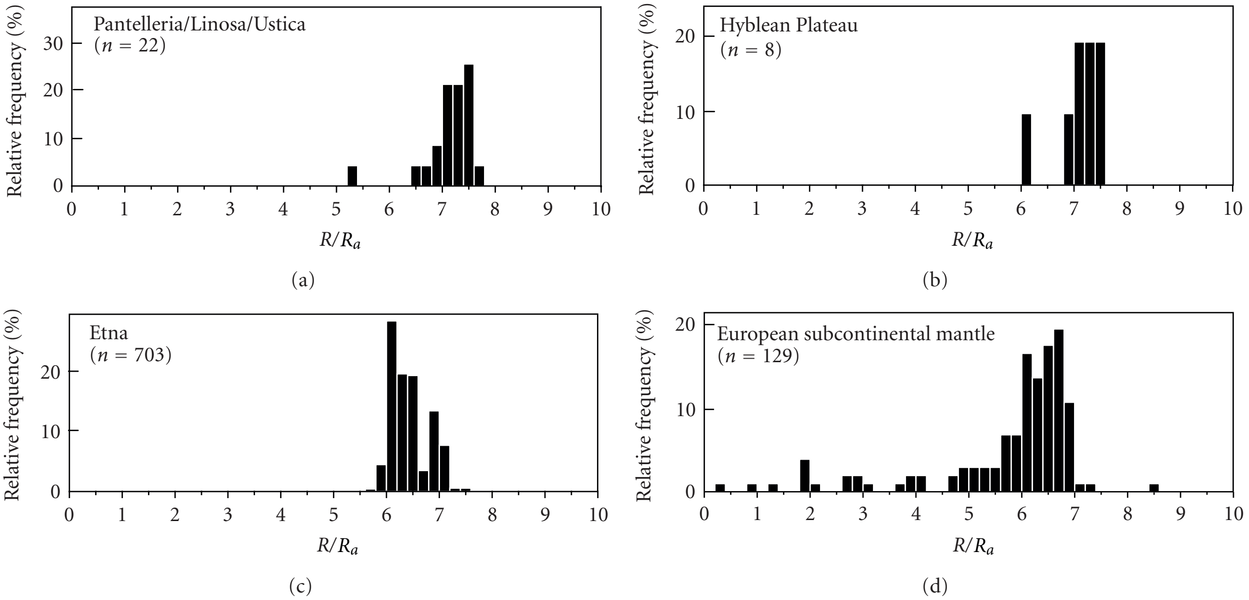
We find that lavas from the three islands display quite homogeneous $^3He/^4He$ values, despite variable total He concentrations. Within experimental uncertainties, Pantelleria lavas ($7.6 \pm 0.3\,R_a$) have a $^3He/^4He$ ratio similar to that measured in the most pristine local volcanic gases ($7.3 \pm 0.1\,R_a$; [24]). Our results for both $^3He/^4He$ and helium abundance in olivine samples from that volcano are in reasonable agreement with those recently published by Martelli et al. [6]: $^3He/^4He$ between 6.95 ± 0.15 and $7.12 \pm 0.3\,R_a$, and $[^4He]$ in the range $(1.9–4.2)\times10^{-9}$ cm^{3}STP/g. We find that Linosa ($7.3 \pm 0.2\,R_a$) but also Ustica ($7.6 \pm 0.2\,R_a$) display $^3He/^4He$ ratios very similar to those of Pantelleria. For Ustica, our $^3He/^4He$ values are actually higher, by as much as one R_a unit, than the values reported by Martelli et al. [6]: 6.5 ± 0.3 to $6.65 \pm 0.2\,R_a$. This may reflect that their olivine samples contained about three orders of magnitude less helium ($2.8–6.9 \times 10^{-10}$ cm^{3}STP/g) than the samples analysed in the present study ($1.6–2.1 \times 10^{-7}$ cm^{3}STP/g).

4. Discussion

Available $^3He/^4He$ ratios for alkaline basaltic volcanoes in southern Italy are summarized in Figure 3. The helium isotope data set for Pantelleria, Linosa, and Ustica indicates a common magmatic helium component feeding the three islands with a $^3He/^4He$ distribution almost identical to that of mantle xenoliths from the Mt Iblei volcanic complex on Sicily mainland [25]. This $^3He/^4He$ range corresponds to the upper range of the $^3He/^4He$ distribution at Mount Etna (Figure 3) and is significantly higher than $^3He/^4He$ ratios of other volcanic series in Italy (Figure 4). $^3He/^4He$ values gradually decrease northward from 2.7 to 7.1 R_a in the Eolian island arc [6, 40–47], 2 to 3.5 R_a at Phlegrean Fields, Ischia and Mount Vesuvius [23, 48–53], down to quite radiogenic values in north-central Italy [54–58].

The uniform He isotopic composition of the three studied islands correlates with the broadly homogenous composition of the erupted mafic magmas in terms of their major and trace element abundances and Sr-Nd-Pb isotopic ratios [1, 8, 11], and references there in; [10, 18]). Even Ustica, which stands on the Tyrrhenian side of the African-European collision plate boundary, significantly differs in both its $^3He/^4He$ ratio and its petrological and geochemical characteristics [6, 17, 59] from calc-alkaline volcanism in the nearby Aeolian Island Arc. It is worth noticing, however, that Alicudi, the westernmost Aeolian Island, displays a $^3He/^4He$ value (6.52–7.07 R_a; [6]) higher than the main Aeolian arc, which may already reflect some OIB-type influence.

Na-alkaline mafic province in southern Mediterranea (Sicily Province), with consistently higher $^3He/^4He$ ratios than any other volcanic series further north in the Italian Peninsula, is tapping a source which is the least contaminated by subduction-related metasomatism, and which may thus represent the mantle end-member of the Italian Plio-Quaternary volcanism. The possible origins of volcanic series in southern Italy have been discussed extensively in the literature with a variety of proposed interpretative models. Reviewing all these models is beyond the scope of this short note (see the recent reviews by [1, 18]), so discussion is focussed on the most salient observations. A key feature derived from seismic studies is the existence of a wide mantle upwelling zone, with low seismic velocity, beneath the whole central-western Europe, eastern Atlantic and the western Mediterranea volcanic provinces [12]. Geochemically, this low velocity component (LVC) appears as a common



(a) (b) (c) (d)

Figure 3: Histogram of helium isotopic ratio measured in volcanic gases and olivines from the islands of Pantelleria, Linosa, and Ustica ([6, 24], and this work), Mts Hyblei ([25], Etna [4–6, 26–28], and Subcontinental Lithospheric Mantle [29–32]).

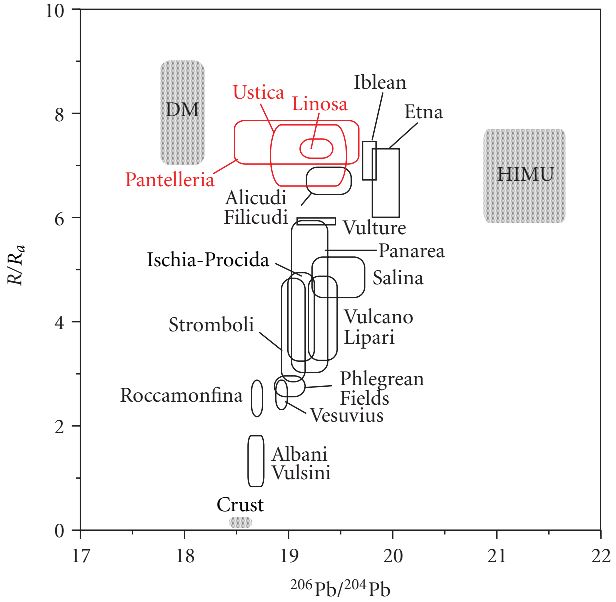


Figure 4: $^3He/^4He$ versus $^{206}Pb/^{204}Pb$ variations for Plio-Quaternary mafic magmas (MgO > 4%) in Italy compared to mantle domains and continental crust: DM stands for Depleted Mantle. The HIMU domain corresponds to the "pure" HIMU component from classical HIMU-type localities such as St. Helena and some Cook-Austral Islands. $^3He/^4He$ range for HIMU is from Hanyu and Kaneoka, (1997) [33] and Hanyu et al., 1999 [34]. All Pb isotopic data and other He data are from the review by Martelli et al, (2008) [6].

sublithospheric mantle end-member to Cenozoic-Quaternary volcanic series in these regions [12]. These rocks display relatively radiogenic Pb and Sr isotopic ratios ($^{206}Pb/^{204}Pb \approx$ 19.9–20.1; $^{87}Sr/^{86}Sr \approx$ 0.7030–0.7034) and exhibit unradiogenic Nd isotopic ratios ($^{143}Nd/^{144}Nd \approx$ 0.51282–0.51294), which point towards a HIMU-like OIB mantle component [54]. Many authors have emphasized this strong HIMU influence in the lavas of the Sicily Province, although this HIMU component may not have the most extreme Pb isotope composition of the classical HIMU endmember measured in OIB from south Pacific localities such as St. Helena and some Cook-Austral Islands [1, 7, 9, 15, 18, 60, 61]. Figure 4 shows the distribution of Italian volcanic series in a $^3He/^4He$-$^{206}Pb/^{204}Pb$ plot, along with the relevant mantle domains and the continental crust. Combining He with lead isotope ratios, rather than Sr-Nd isotope ratios, provides more discriminating information on possible mantle sources of basaltic volcanism, as Pb is a more incompatible element during magmatic processes and a more sensitive tracer of lithosphere recycling into the mantle. The spectacular downward trend for lavas from the Aeolian Island Arc and the Italian Peninsula in Figure 4 typically depicts the northward increasing contamination of the mantle source by Pb-rich crustal material derived from either subducted slabs or continental crust. ^{206}Pb/^{204}Pb ratios (and also Sr and Nd isotope ratios), for Etna lavas have isotopic characteristics that are identical to the LVC end-member even though these lavas also display some trace element ratios indicative of a subduction influence. In turn ^{3}He/^{4}He distributions for Etna and for the Subcontinental Lithospheric Mantle are nearly indistinguishable statistically (Figure 3), thus indicating that the isotopic similarity between Etna and LVC also apply to helium isotopes. In contrast, Pantelleria, Linosa, and Ustica volcanic islands display less radiogenic ratios and plot in intermediate position between this end-member and the Depleted Mantle (MORB mantle) domain, suggesting that additional ambient MORB asthenosphere may be entrained at the periphery of mantle upwelling.

5. Conclusions

The presented helium isotope measurements in olivines crystals from Ustica, Linosa and Pantelleria volcanic islands indicate the following.

(i) All three islands display quite homogeneous helium isotope ratios, and ^{3}He/^{4}He values for olivine crystals are concordant with ratios in present-day volcanic gases of Pantelleria.

(ii) Based on all available data, the ^{3}He/^{4}He range delineates a quite homogeneous 250 km-wide magmatic province with a ^{3}He/^{4}He ratio higher than anywhere else further north in Italy.

(iii) Consistent with previous studies [4, 6, 12, 18, 25], the He-Pb isotope systematics show that this magmatic province may be fed by a mixture of HIMU-type mantle and MORB asthenosphere. These results are consistent with little or no contribution from a third ^{3}He-richer component, in agreement with the MORB-type neon isotopic composition in Etna lavas [62] and with seismic inferences of an upwelling (“plume”) mantle source that is prevalently rooted near the transition zone at 660 km depth [12, 13].

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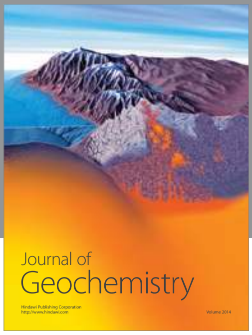
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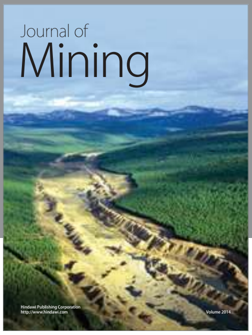
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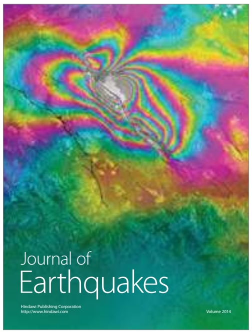
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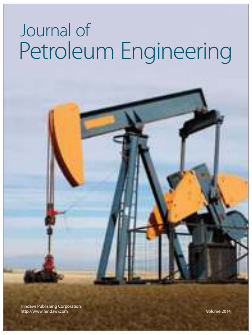
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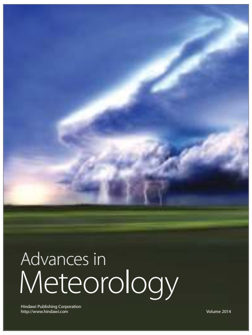
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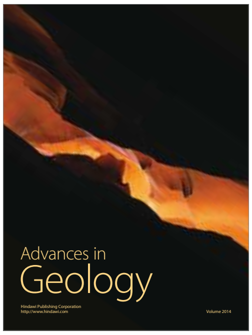
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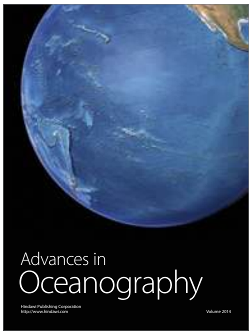
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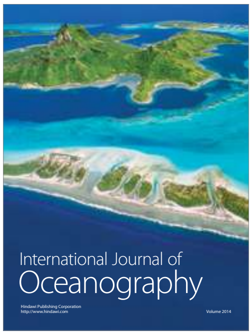
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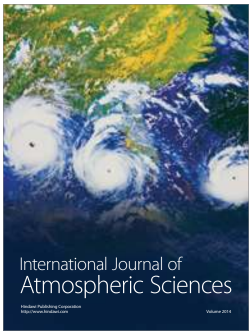
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