Constraints on mantle source and interactions from He-Sr isotope variation in Italian Plio-Quaternary volcanism

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Abstract

Helium isotope ratios of olivine and pyroxene phenocrysts from Plio-Quaternary volcanic rocks from Southern Italy (seven Eolian Islands, Mt. Vulture, Etna, Ustica, Pantelleria) range from 2.3 to 7.1 R_a. Importantly the phenocryst $^3He/^4He$ correlate well with whole rock Sr isotopic composition (0.70309-0.70711) reflecting the mixing of two sources. A significant contribution of He from crustal contamination is recorded only occasionally (e.g., pyroxenes from Vulcano). When merged with data from the Roman Comagmatic Province, a remarkably strong near-linear He-Sr isotope correlation is apparent. The general northward decrease in $^3He/^4He$ corresponds to an increase in $^{87}Sr/^{86}Sr$ (and decrease in $^{143}Nd/^{144}Nd$ and $^{206}Pb/^{204}Pb$) that is due to increasing metasomatic enrichment of the mantle wedge via subduction of the Ionian-Adriatic plate. Calculations based on the ingrowth of ^4He in the wedge and on the ^4He content of the subducting crust show that mechanisms of enrichment in radiogenic He are effective only if the wedge is strongly depleted in He relative to best estimates of the depleted mantle. This can be accommodated if the process of metasomatism by the subduction fluids depletes the mantle wedge. The $^3He/^4He$ of Pantelleria, Etna, Iblei, Ustica, Alicudi and Filicudi basalts (7.0 ± 0.6 R_a) define the mantle composition least affected by subduction-related metasomatism. Although these volcanoes are from a variety of tectonic regimes (subduction-related, intraplate, rifting) their similarities suggest a common origin of geochemical features. Their characteristics are consistent with a HIMU-type mantle that is either younger than the Cook-Austral island end-member, or has a lower $^{238}U/^{204}Pb$.

1. Introduction

Helium isotopes in arc basalts trace the contribution of mantle- and crust-derived volatiles in the generation of melts at subduction zones. The ^{3}He/^{4}He of oceanic arc basalts are commonly in the range 6-8 R_a (where R_a is the atmospheric ^{3}He/^{4}He; 1.39 x 10^{-6}) [*Poreda and Craig*, 1989]; values that are typical of normal mid-ocean ridge basalts (MORB) are in the range of 8 ±1 R_a [*Farley and Neroda*, 1998]. Helium in crustal fluids is enriched in radiogenic ^{4}He produced by the decay of U and Th. Crustal-radiogenic He is typically less then 0.1 R_a [*O'Nions and Oxburgh*, 1988], the large difference from mantle values make He isotopes a powerful tracer of crust-derived volatiles in magmatic systems. The absence of a significant contribution of radiogenic He in oceanic arc basalts implies that the subduction of altered oceanic crust and oceanic sediments does not enrich the mantle wedge in radiogenic helium. This is likely due to the loss of He from the down-going slab in the early stages of subduction prior to reaching the zone of magma generation in the mantle wedge [*Hilton et al.*, 1992, 2002].

Crustal contamination of arc magmas erupted through continental crust is common and is typically reflected in the low ^{3}He/^{4}He of pyroxene phenocrysts compared to cogenetic olivine [*e.g. Hilton et al.*, 1993a,b]. The presence of radiogenic helium in the mantle wedge source region of basalts is only recorded in two arcs: the east Sunda-Banda arc, Indonesia [*Hilton et al*., 1992] and the Roman Comagmatic Province (RCP) of central-northern Italy [*Martelli et al*., 2004]. At both arcs continental crust (or continent-derived sediment) is currently being subducted and it is tempting to assume that crustal-radiogenic helium has been recycled into the mantle wedge. This would require models of global He isotope systematics to be reconsidered.

In our previous work on the Roman Comagmatic Province (Latium and Campania regions) we showed that the basaltic rocks display a coherent correlation between the He and Sr isotope compositions that implied the two elements are strongly coupled during the subduction process [*Martelli et al*., 2004]. However, it was unclear whether the low ^{3}He/^{4}He resulted from post-metasomatic radiogenic ingrowth in a He-poor mantle or addition of radiogenic He from subducting crust. Here we complete the systematic survey of the He isotopes in Italian Plio-Quaternary basalts by reporting new data from the volcanic provinces of southern Italy; specifically the Vulture volcanic region, the Aeolian islands, Ustica, Etna and Pantelleria. The helium isotope data are combined with new Sr and Pb isotope determinations in order to constrain the characteristics of the sub-Italian mantle and the source of radiogenic helium in mantle wedge.

2 Geochemistry and geodynamic setting of south Italy volcanism

The Plio-Quaternary mafic volcanic rocks of the Italian peninsula (Figure 1) display extreme petrologic and geochemical variation. Volcanic rocks with more than 4% MgO have K_2O/Na_2O that range from 0.1 to 10 [*Peccerillo*, 2005]. Calc-alkaline to shoshonitic rocks dominate the Aeolian Islands, Na-alkaline at Etna, Ustica and the Iblean plateau, K-alkaline and calc-alkaline rocks are typical of the RCP, while basalts from the Sicily Channel are alkaline [*Ellam et al*., 1989; *Wilson and Bianchini*, 1999]. These differences are largely a function of the complex geodynamic history of the region over the last 300 Myr [*e.g. Peccerillo and Turco*, 2004]. The Plio-Quaternary volcanism reflects the latest part of this process, and is in large part due to the subduction of the Ionian-Adriatic plate in the last 25-30 Myr [*Doglioni et al*., 1999].

The Aeolian volcanic arc has been generated by melt production in the mantle above the westward subduction of the Ionian plate [*Barberi et al.*, 1973]. Volcanic activity dates back to at least 600 ka, and active volcanism occurs today at Vulcano, Stromboli and Panarea. Although there is ample evidence that the magmas underwent interaction with crustal rocks [e.g. *Ellam et al.*, 1989; *De Astis et al*., 2000], regional-scale isotopic and trace variations are difficult to explain by assimilation, and unacceptably large degrees of contamination are often required to account for the Sr-Nd-Pb isotopic composition of many basalts [e.g. Stromboli; *De Astis et al*., 2000]. The Eolian islands display a large range in ratios of large ion lithophile elements (LILE) over high field strength elements (HFSE) and Sr, Nd and Pb isotopic compositions. From west (Alicudi and Filicudi) to east (Stromboli) (Figure 1) potassic basalts become relatively more important than calc-alkaline basalts, Sr isotope ratios and LILE/HFSE ratios increase, and Nd and Pb isotope ratios decrease [*Calanchi et al.*, 2002]. These major geochemical changes reflect the heterogeneties in the sub-Italian mantle [*Peccerillo*, 2005]. It is well established that the mantle heterogeneity has been produced by fluids released by the subducting Ionian-Adriatic plate over the last 30 million years [*Civetta et al*., 1981; *Beccaluva et al.*, 1991; *Peccerillo*, 1999; *Wilson and Bianchini*, 1999]. The ultimate origin of the fluids that have metasomatised the sub-Italian mantle is, however, not well established. Several hypotheses have been developed to explain the observed trace elemental and isotopic composition variation in south Italian basalts: melts produced by continental sediments [*Beccaluva et al.*, 1991], aqueous fluids [*De Astis et al.*, 2000; *Santo et al.*, 2004], aqueous fluids plus silicate melts [*Wilson and Bianchini*, 1999], carbonate plus silicate component [*Conticelli et al.*, 2004].

Monte Vulture is an isolated stratovolcanic centre in south central Italy, east of the Roman and Campanian alignment (Figure 1). It is located at the outer front of the Apennine orogen at the edge of the Apulian foreland [*Beccaluva et al.*, 2002]. Magmatism is dominated by Na-K-rich tephrites and phonolites which were erupted between 800 and 100 ka [*Peccerillo*, 2005]. Like the volcanic rocks of the Tyrrhenian margin the Monte Vulture volcanics have high LFSE/HFSE and negative Ta, Nb and Ti anomalies, which are attributed to a subduction origin. However, basaltic flows from Monte Vulture have lower Th/Nb and distinctive LREE and P enrichment which is argued to reflect a contribution from intraplate magmatism [*Beccaluva et al.*, 2002]. Recent studies suggest that Monte Vulture sits above a region where the subducting slab has become detached, permitting sub-African asthenospheric mantle to mix with sub-Tyrrhenian mantle. The sub-Tyrrhenian mantle was previously metasomatised by subduction-related fluids [*De Astis et al.*, 2006].

The basaltic volcanism of Etna and Ustica is distinct from most of the south Italian arc volcanism. Both centres appear to be related to NW-SE faulting in the subducting Adriatic plate that has driven upward flow of mantle melts [*Gvirtzman and Nur*, 1999; *Doglioni et al.*, 2001; *Trua et al.*, 2003]. Na-alkaline magmatism dominates at Etna. Both sub-alkaline and alkaline basalts were erupted at Ustica from 750 to 130 ka. The Iblean plateau is the foreland of the Apennine subduction and has not been involved in subduction processes. The Iblean basalts, basaltic andesites and nephelinites, with sodic alkaline and sub-alkaline affinity, were erupted between 7.5 and 1.5 Ma.

Pantelleria island is located approximately 100 km south-west of Sicily in the Sicily Channel. It is situated on a NW-SE trending rift that appears to be the result of trans-tensional tectonics along the northern margin of the African Plate [*Boccaletti et al.*, 1987]. Mafic magmas are transitional- to weakly-alkaline and were erupted between 300 and 5 ka [*Peccerillo*, 2005]. Trace element ratios (e.g. Ta/Yb, Th/Yb, Nb/Zr) have intraplate characteristics [*Wilson and Bianchini*, 1999].

3. Samples and analytical procedures

To determine the He isotope composition of the mantle beneath southern Italy, fresh olivine and/or pyroxene phenocryst-bearing basaltic lavas or pyroclastic deposits were sampled from the each of the Aeolian islands, Ustica, Pantelleria, Vulture and Etna (Figure 1). Details of sample location and rock-type are given in Table 1. To avoid cosmogenic ^{3}He contamination most samples are from road-cuts or rapidly eroding slopes. Samples are less than 210 ka and most have ^{3}He/^{4}He that are higher than similar aged basalts from the Campania-Latium regions of the RCP where a significant contribution of radiogenic ^{4}He was excluded [*Martelli et al.*, 2004]. These observations suggest that even for new samples we can exclude massive presence of radiogenic ^{4}He.

Helium isotopes were measured in gases released by *in vacuo* crushing of olivine and pyroxene phenocryst separates using procedures similar to *Stuart et al.* [2000]. The hydraulic crusher used in this study does not release lattice-hosted radiogenic [*Stuart et al.*, 2003] or cosmogenic He [*Williams et al.*, 2005]. Strontium isotopes were measured on powdered basalt whole rock samples used for helium isotope measurement, or on the powders of pyroxene from the pyroclastic rocks that remained after *in vacuo* crush extraction of helium [*Martelli et al.*, 2004]. Lead isotope determinations on three basalt samples from Pantelleria used the procedures of *Ellam* [2006].

4. Results

The ^{3}He/^{4}He and ^{87}Sr/^{86}Sr values of south Italy basalts are presented in Table 1 and Figures 2 and 3. The phenocryst ^{3}He/^{4}He ratios range from 2.3 to 7.1 R_a which overlaps and extends to higher values the range recorded by basaltic rocks from the Campanian and Roman provinces [0.44-5.2 R_a, *Martelli et al.*, 2004] (Figure 3). The most radiogenic ^{3}He/^{4}He ratios are recorded by Strombolian basalts (2.7-4.8 R_a). These values overlap the ^{3}He/^{4}He of the Campanian province as recorded by basalts from Procida and Vesuvius [2.5-5.2 R_a, *Martelli et al.*, 2004]. The highest ^{3}He/^{4}He of Strombolian basalts overlap values recorded by flows from Salina, Lipari and Vulcano (4.5-5.5 R_a).

The two Panarea samples display very different ^{3}He/^{4}He (La Fossa; ~3 R_a and Punta Torrione; ~6 R_a). These flows belong to different volcanic series that are interpreted to be derived from different mantle sources. The Punta Torrione flow belongs to the calc-alkaline basalt series that are similar in composition to the western arc, while La Fossa is geochemically similar to Stromboli [*Calanchi et al.*, 2002]. The separate sources are also reflected in the different ^{87}Sr/^{86}Sr of the La Fossa (0.7053) and Punta Torrione flows (0.7046).

The western-most Aeolian islands (Alicudi and Filicudi) have the highest $^3He/^4He$ (6.7-7.1 R_a). These are similar to the values measured in olivine-bearing basalt flows from Ustica (~6.6 R_a) and Etna [6.3-7.1 R_a; *Marty et al.*, 1994, this work]. Olivine phenocrysts from three alkali basalt flows from Pantelleria have similarly high $^3He/^4He$ (7 R_a). Olivine from a pyroclastic surge and a xenolith from Monte Vulture have $^3He/^4He$ of ~6 R_a.

In general the olivine $^3He/^4He$ are consistent with values of magmatic gases and aqueous fluids from each volcanic centre (see Figure 4 for detailed description), confirming that the fluids' maximum $^3He/^4He$ generally reflects the degassing of magmatic bodies at depth [*Martelli et al.*, 2004]. In the case of Vulcano, the fumarole $^3He/^4He$ are greater than the phenocryst $^3He/^4He$ (Figure 4). Similar features have been observed at Cerro Negro [Nicaragua, *Fisher et al.*, 1999], Canary Islands [*Hilton et al.*, 2000] and Etna [*Rizzo et al.*, 2006]. This may reflect subtle temporal changes in He isotopic composition of the mantle source or crustal contamination [*Hilton et al.*, 1993a, b], or the fractionation of magmatic He isotopes in fumarole gases [*Rizzo et al.*, 2006].

Whole rock $^{87}Sr/^{86}Sr$ range from 0.70308 to 0.7071 (Table 1, Figure 2). This overlaps with the range recorded by basaltic rocks from the Campanian and Roman provinces [*Martelli et al.*, 2004] (see Figure 3), but extends to lower values. Aeolian island basalts display nearly the complete range of $^{87}Sr/^{86}Sr$: from 0.70367 at Alicudi to 0.7071 at Stromboli. These values are indistinguishable from previous determinations of Aeolian basalts [*Calanchi et al.*, 2002 and references therein]. The Monte Vulture basalt $^{87}Sr/^{86}Sr$ (0.70564) falls within the range recorded in a more extensive study by *De Astis et al*. [2006]. The most unradiogenic Sr isotope ratios are recorded by basalts from Pantelleria (0.70308-0.70311) and Ustica (0.70320-0.70332), which are again similar to previous measurements [*Civetta et al.*, 1998; *Trua et al.*, 2003].

5. Discussion

5.1 He-Sr isotope systematics of Italian Plio-Quaternary volcanism

It is widely accepted that much of the geochemical variation of Italian Plio-Quaternary volcanism reflects variation in mantle composition [e.g. *De Astis et al.*, 2000; *Gasperini et al.*, 2002; *Peccerillo and Lustrino*, 2005]. Trace element and Sr, Nd and Pb isotope variation demonstrate a progressive northward mantle enrichment [e.g. *Gasperini et al.*, 2002]. A correlation trend in He-Sr isotope space defined by the Roman Province basalts has previously been interpreted as a binary mix between a high $^3He/^4He$-low $^{87}Sr/^{86}Sr$

asthenospheric mantle source and a low $^{3}He/^{4}He$-high $^{87}Sr/^{86}Sr$ component consistent with metasomatically-altered mantle [*Martelli et al.*, 2004]. The new He-Sr isotope data from southern Italy continue this trend to higher $^{3}He/^{4}He$ and lower $^{87}Sr/^{86}Sr$, re-affirming the general trend of a southward $^{87}Sr/^{86}Sr$ decrease that is associated with increasing $^{3}He/^{4}He$ (Figure 3). The high $^{3}He/^{4}He$-low $^{87}Sr/^{86}Sr$ end of the array is defined by Pantelleria, Ustica, Etna and the western Aeolian islands (Alicudi and Filicudi). The $^{3}He/^{4}He$ (6.7-7.1 R_a) and $^{87}Sr/^{86}Sr$ (0.7030-0.7036) overlap published values of basalts from the Iblean plateau [*Trua et al.*, 1998; *Sapienza et al.*, 2005] and represent the best estimate of mantle uncontaminated by fluids from subducting crust and/or sediments.

Basalts with $^{3}He/^{4}He$ lower than the south Italy maximum (~7 R_a) contain radiogenic He. In the prevailing hypothesis this is derived from the mantle wedge [*Martelli et al.*, 2004]. However, crustal contamination of magmas prior to eruption is recorded occasionally in south Italian volcanism [*Ellam and Harmon*, 1990; *De Astis et al.*, 2000]. Of the eight co-genetic olivine and pyroxene phenocrysts (Table 1), the pyroxene from three samples have lower $^{3}He/^{4}He$ than the olivine. This could be indicative of subtle crustal contamination and consequently pyroxene $^{3}He/^{4}He$ measurements can be only considered a lower limit on the magmatic value. It is notable that basalts from Vulcano display a range of $^{3}He/^{4}He$ (3.3 to 4.9 R_a) that is not reflected in a concomitant change in $^{87}Sr/^{86}Sr$ (Figure 2). This probably originates from the addition of crustal-radiogenic He to a He-poor magma, probably due to shallow degassing [*Hilton et al.*, 1993b]. The $^{87}Sr/^{86}Sr$ of the basalts is less sensitive to crustal contamination as the Sr concentration of the basaltic melts is higher than in the contaminating crust [*Ellam and Harmon*, 1990]. Similar conclusions were proposed by *Ellam and Harmon* [1990] based on the Sr-O systematics in the Aeolian lavas.

The Plio-Quaternary Italian basalts appear to define a near-linear trend in He-Sr isotope space (Figure 3). This coherent relationship between He and a lithophile radiogenic isotope tracer is rare. Its occurrence over a large part of $^{87}Sr/^{86}Sr$ observed in mantle rocks argues strongly against it being fortuitous. The lowest $^{87}Sr/^{86}Sr$ are consistent with previous determinations of uncontaminated sub-Italian mantle (0.7025) [e.g., *Gasperini et al.* 2002]. However, there is considerable uncertainty in the $^{87}Sr/^{86}Sr$ of the crustal component due in large part to the apparently high $^{87}Sr/^{86}Sr$ of the most enriched Tuscan lamproites [~0.715; *Conticelli and Peccerillo*, 1992; *Gasperini et al.*, 2002]. It is frequently argued that the Tuscan volcanic rocks involve a third "crustal" component with more radiogenic Sr than is present elsewhere in Italian Plio-Quaternary volcanism [e.g. *Rogers et al.* 1985; *Ellam et al.* 1989; *Gasperini et al.*, 2002]. The well-defined isotopic composition of crustal He can be combined with the coherent He-Sr isotope relationship (Figure 3) to establish the $^{87}Sr/^{86}Sr$ of the crustal component. Simply extrapolating a linear fit to an end-member with $^{3}He/^{4}He < 0.1\ R_a$ (undiluted radiogenic He) implies that the crustal component has $^{87}Sr/^{86}Sr \sim 0.712$ (Figure 3). One implication of the essentially linear $^{3}He/^{4}He$-$^{87}Sr/^{86}Sr$ trend exhibited by the data (Figure 3) is that the (He/Sr) of the crustal component mantle must be similar to the (He/Sr) of the unaltered mantle in the wedge ($K = (He/Sr)_c/(He/Sr)_m$ ranges between 1 and 4; c: crust, m: mantle). In Figure 5 we have plotted compilations of He-Nd and He-Pb isotope measurements from Italian basalts (see figure caption for details). Although the data were not measured on the same samples they appear to define binary mixing trends similar to the He-Sr isotope trends. For a mantle component with $^{143}Nd/^{144}Nd = 0.51305$ and $^{206}Pb/^{204}Pb = 19.9$, and crustal component with $^{143}Nd/^{144}Nd = 0.51205$ and $^{206}Pb/^{204}Pb = 18.55$, $K_{He\text{-}Nd}$ and $K_{He\text{-}Pb}$ range between 0.3 and 3 (Figure 5). In this case the K values do not change significantly if Tuscan lamproites are considered because their $^{143}Nd/^{144}Nd$ and $^{206}Pb/^{204}Pb$ are similar to Latium basalts [*Peccerillo*, 2005]. This result is consistent with the near-linear or slightly curved Sr-Nd and Sr-Pb isotope trends of Italian basalts without inclusion of the Tuscan rocks [e.g. *Gasperini et al.,* 2002].

5.2 The origin of radiogenic He in Italian Plio-Quaternary volcanism

The low helium isotope ratios of basalts from the eastern Aeolian islands and mainland Italy [down to ~ 1 R_a, *Martelli et al.*, 2004] are atypical of subduction zones. The absence of radiogenic He in oceanic arc basalts and fluids suggests that the direct addition of radiogenic He by subduction of oceanic lithosphere is unlikely [*Patterson et al.*, 1994; *Dodson and Brandon*, 1999; *Bach and Niedermann*, 1998; *Hilton et al.*, 2002]. The presence of radiogenic He in the Plio-Quaternary basalts of Italy, therefore, relies on a continental source. The presence of subducted continental crust in Italy is well documented [e.g. *Carminati et al.*, 2005]. The Banda arc is the only other subduction zone where low $^{3}He/^{4}He$ basalts result from interaction of the mantle wedge with the subducting continental crust [*Hilton et al.*, 1992]. In contrast to Italy, the He isotope composition of Banda arc basalts appears not to be strongly coupled to Sr isotopes and other petrogenetic tracers [*Hilton et al.,* 1992]. The low $^{3}He/^{4}He$ of the Banda arc basalts has been attributed to contamination of the mantle wedge by subduction of continental crust [*Hilton et al.,* 1992]. However, the isotopic-trace element signature of Italian arc basalts supports contamination of the mantle wedge by fluids derived from subducted crustal rocks [*Gasperini et al.,* 2002; *Peccerillo,* 2005]. A sharp change in

$^3He/^4He$ at Banda (from 6 to 1.2 R_a) over a 70 km-wide zone is interpreted as a transition between the subduction of oceanic and continental slabs [*Hilton and Craig,* 1989]. The existence of an oceanic-continental crust transition in Italy is unclear [e.g. *Amato and Montone,* 1997] and the $^3He/^4He$ distribution shows no sharp change consistent with a net transition.

The radiogenic He in the mantle wedge may originate from two sources; (i) the ^4He ingrowth in the mantle wedge after metasomatic enrichment of U and Th, and/or (ii) addition of crustal-radiogenic He to the mantle wedge via fluids from the subducted slab.

The effect that both processes have on altering the isotopic composition of mantle He depends strongly on the initial concentration of the unmodified mantle. This is an admittedly poorly-constrained parameter. The depleted MORB-mantle (DMM) source has a relatively well-established He concentration (1.5 x 10^{-5} cc STP/g) [*Allegre et al.*, 1986-87; *Sarda and Graham,* 1990]. In common with other studies [e.g. *Dunai and Baur*, 1995; *Hilton et al.*, 2000; *Shaw et al.*, 2006] in the following discussion we use DMM He concentrations for illustrative purposes, although it should be borne in mind that the sub-Italian mantle may have a slightly different He concentration.

(i) The duration of the ingrowth of ^4He due to metasomatic addition of U and Th from the slab is limited. Westward subduction of the Ionian-Adriatic plate started no earlier than 30 Ma [*Doglioni et al.* 1999] and provides an upper limit for the duration of ingrowth. The metasomatised mantle has a maximum content of 200 ppm U and 950 ppm Th and this produces 1.5 x10^{-6} ccSTP ^4He /g in 30 Myr [*Martelli et al.,* 2004]. This is sufficient to lower the $^3He/^4He$ of a DMM source (typically 7-9 R_a) by less than 10% and cannot explain the low ratios of Italian basalts. Post-metasomatic He ingrowth can only decrease mantle $^3He/^4He$ significantly if the initial mantle He concentration is two orders of magnitude or less than DMM concentration.

(ii) For the radiogenic He in the Italian basalts to originate in subducted continent-derived material we require a mechanism to transport the crustal He to the fluids that metasomatise the mantle wedge. Of the common rock-forming minerals, garnet likely has the highest closure temperature [T_c = 600°C, *Dunai and Roselieb,* 1996] and will transport the crustal-radiogenic He to the greatest depth. It is worth noting that if T_c of He in garnet is as low as proposed by *Blackburn and Stockli* [2006] (110-300°C), He would not be transported at significant depth. In order to estimate the maximum amount of He that could be transferred to the mantle wedge above the subducting slab, we assume that all the He produced in the garnet of the crustal basement is entirely transferred to the wedge via an aqueous fluid or melt (Table 3, Figure 6). Using the parameters in Table 3 we estimate that the Adriatic basement has approximately 1.1 x 10^{-6} cc ^{4}He STP/g. The effect that the subduction of this He has on decreasing the mantle ^{3}He/^{4}He ratios depends the volume of mantle that it affects. Figure 7 illustrates that, assuming an initial $[He]_{DMM}$, the addition of crustal He decreases the ^{3}He/^{4}He to 1 R_a only if the volume of metasomatised mantle is very small (E-W extension = 7 km). This is not compatible with the geographic distribution of the contaminated wedge as we can assume that the E-W extension should be at least 45 km (i.e., the distance from Vesuvio to Ischia in Campania region, both contaminated by the subduction). In order to satisfy this constraint, the initial [He] of the mantle should be lower than 6 x 10^{-7} cc STP/g.

Therefore, neither post-metasomatic ingrowth nor the direct addition of crustal He can explain the low ^{3}He/^{4}He mantle that is prevalent in the Italian magmatism if it starts with DMM He concentrations. A mantle reservoir with He concentration low enough for either mechanism to have generated the radiogenic ^{3}He/^{4}He would rapidly evolve low ^{3}He/^{4}He (unless buffered by the addition He from elsewhere). It is highly unlikely that a low [He] HIMU mantle reservoir could consistently evolve the remarkably constant ^{3}He/^{4}He that is typical of the global HIMU mantle-source [*Hanyu and Kaneoka*, 1997]. Instead, it is more likely that mantle He is lost as a result of the process of metasomatism. One mechanism may be that the percolation of the metasomatic fluid devolatilises the mantle wedge in a manner similar to that during aqueous/carbonic fluid infiltration through crustal rocks [*Bickle and Baker*, 1990]. Studies of incipient charnockite formation in southern India, for instance, suggest that a dehydration reaction front propagates through the silicate rock due to advection of an infiltrating CO_2-rich fluid [*Harris and Bickle*, 1989]. This process tends to remove the soluble components, including the inert gases, leaving behind a U- and Th-rich but He-poor mantle that is susceptible to radiogenic ingrowth. Although we cannot rule out the possibility that a proportion of the radiogenic He in the sub-Italian mantle is derived from the infiltrating metasomatic fluids, post-metasomatic ingrowth can account for the low ^{3}He/^{4}He and coherent He-Sr isotope trend if the mantle wedge was sufficiently degassed during metasomatism.

5.3 Mantle source of south Italian volcanism

The He isotopic composition of Pantelleria, Alicudi, Filicudi, Ustica and Etna (6.7-7.1 R_a) are the highest in the region and imply that crustal fluids have not significantly modified the

He isotopes of the mantle source. This range overlaps values of xenoliths from Iblean plateau basalts [7.3 ± 0.3 R_a; *Sapienza et al.*, 2005]. Anyway, if we consider the Sr, Nd and Pb isotopes, the uncontaminated mantle end member should be restricted to Etna, Iblei and Pantelleria (Figure 3 and 5).

In plots of Sr vs. Pb and Nd vs. Pb isotopes the most primitive (i.e., not contaminated by the subduction) Italian volcanism lies intermediate between compositions that are typical of sub-oceanic depleted mantle (DMM) and HIMU-type mantle (high μ = high $^{238}U/^{204}Pb$) [*Civetta et al.*, 1998; *Gasperini et al.*, 2002]. Some authors [e.g. *Peccerillo*, 2003; *De Astis et al.*, 2006] have argued that the relatively depleted mantle melts in southern Italy are compositionally similar to the deep mantle proposed to originate in the so-called Focal-zone (FOZO) of ocean island basalts [e.g. *Hart et al.* 1992]. Hart and co-workers argue that the FOZO mantle is the source of high $^3He/^4He$ in ocean island basalts. If this is correct, this implies that the Italian volcanism should have $^3He/^4He$ higher than typical values of MORB, conceivably up to 50 R_a [*Stuart et al.*, 2003]. Although there is abundant evidence that the high $^3He/^4He$ mantle has a composition similar to depleted mantle [e.g. *Stuart et al.*, 2003], the low $^3He/^4He$ of the uncontaminated basalts clearly rules out FOZO mantle beneath southern Italy and the Sicily Channel. The average $^3He/^4He$ of olivine from Alicudi, Filicudi, Ustica, Etna, Iblei and Pantelleria is 6.8 ± 0.2 R_a. Although this does not unequivocally rule out a depleted mantle source [MORB: 8 ± 1 R_a; *Farley and Neroda*, 1998] it is remarkably similar to the average $^3He/^4He$ of the HIMU end-member [6.8 ± 0.9 R_a; *Hanyu and Kaneoka*, 1997, *Moreira and Kurz*, 2001] as defined by basalts with $^{206}Pb/^{204}Pb > 20.3$.

Also, the presence of the EM1-type enriched mantle [$^{87}Sr/^{86}Sr \sim 0.705$ and $^{143}Nd/^{144}Nd \sim 0.51245$; *Zindler and Hart*, 1986, *Hofmann*, 1997] has been suggested to explain the isotopic composition of basalts from Alicudi [*Peccerillo et al.*, 2004] and Pantelleria [*Civetta et al.*, 1998]. $^3He/^4He$ of EM1-type basalts is not precisely determined [*Hanan and Graham*, 1996; *Eiler et al.*, 1997] and consequently cannot be used to distinguish a contribution. The Pb isotope composition of the Pantelleria basalts ($^{206}Pb/^{204}Pb$ = 19.32-19.67; Table 2) overlaps the range previously measured [$^{206}Pb/^{204}Pb$ =19.09-19.69; *Esperanca and Crisci*, 1995] and is significantly more radiogenic than EM1 [$^{206}Pb/^{204}Pb$ = 17.0-18.5; *Hofmann*, 1997], clearly indicative of HIMU-type mantle.

The maximum $^{143}Nd/^{144}Nd$ and $^{206}Pb/^{204}Pb$ measured in basalts from Etna, Iblei and Pantelleria are between 0.51290 and 0.51302 and 19.8 and 20.0, respectively [*Marty et al.*, 1994; *Civetta et al.*, 1998; *Gasperini et al.*, 2002]. Such values are in the range of the Low Velocity Composition, as defined by Hoernle et al. (1995), a uniform geochemical reservoir that beside southern Italy is tapped by volcanism of the Canary islands and Central European volcanic province (Massif Central, Eifel, Rhine graben, Lower Silesia, western Pannonian basin). The magmatism of this province has also been included into the Common Mantle Reservoir (CMR), a widespread igneous province developed within the Mediterranean sea and surrounding regions [*Lustrino and Wilson*, 2007]. Importantly, helium isotopes in the European Cenozoic Provinces [*Dunai and Baur*, 1995; *Gautheron et al.* 2005], and north Africa [*Barfod et al.*, 1999; *Beccaluva et al.* 2007a,b] are indistinguishable from the south Italy average. These regions have very different lithospheric history and tectonic regimes. For instance, Pantelleria is associated with lithospheric rifting, Iblei is a foreland region and Etna is a mixed intraplate-subduction volcanism [*Schiano et al., 2001; Tonarini et al., 2001*]. The near-identical Sr, Nd, Pb and He isotope geochemistry of the volcanics favours a regionally common mantle origin over local explanations. For instance, it is difficult to reconcile an origin for the south Italian mantle composition in locally upwelling deep mantle [*Gasperini et al.*, 2002] and in absence of geophysical evidence for the necessary slab window [e.g. *Lucente et al.*, 1999]. A common source for the European/African volcanism has been related to the broad upwelling of a deep mantle plume based on geophysical observations [*Hoernle et al.*, 1995]. It is well-established that the enriched trace element composition and isotope geochemistry of HIMU-like mantle can be generated by the storage of small volume alkali- and volatile-rich melts at the base of the oceanic and continental lithosphere for a few 100 million years [*Halliday et al.*, 1995; *Niu and O'Hara*, 2003; *Panter et al*., 2006]. Recent geochemical studies of xenolith suites from the Europe and North Africa have shown that the HIMU signature has been generated by enrichment of lithosphere by fluids and melts [*e.g. Pilet et al.*, 2005; *Beccaluva et al*., 2007b].

The Pb isotope composition of south Italian basalts are less radiogenic than the Cook-Austral islands and St. Helena basalts that define the HIMU end-member [*Chaffey et al*., 1989; *Woodhead et al.*, 1996]. Carbonatite metasomatism has been proposed for the origin of the HIMU characteristics [*Hauri et al.*, 1993]. This hypothesis is not fully supported by the geochemistry of the Italian basalts. The anomalies evident in the trace-elements patterns of carbonatite, in particular high Zr/Hf [~60, *Chakhmouradian* 2006], are not observed in the Italian basalts. For example, using the database of Peccerillo (2005), the average Zr/Hf of Etna, Iblean and Pantelleria basalts is 46 (n = 298). The standard model predicts an origin for the south Italian mantle as a mix between HIMU and a pre-existing depleted mantle end-members [*e.g. Civetta et al.*, 1998; *Gasperini et al*., 2002]. However, the similarity of the Pb, Sr, Nd and He isotope composition of intraplate volcanism across Europe and North Africa is difficult to reconcile with a two component mixing process that, by it's nature, will vary spatially and temporally. It seems more likely that the HIMU-like signature reflects a single mantle composition that has either not had as long as the Cook-Austral island source to grow-in radiogenic Pb, or has a lower $^{238}U/^{204}Pb$. The absence of depleted mantle is supported by the incompatible trace element signature of melt inclusions in olivine phenocrysts from Ustica, Etna and Iblean plateau that are remarkably similar to typical HIMU [*Schiano et al*., 2004]. Therefore the alternative explanation is that the south Italy mantle end-member has been generated by enrichment of the lithosphere by alkaline carbonate-rich melts. In this case passive asthenospheric mantle uprising and decompression melting is linked to tensional stresses in the lithosphere during Cenozoic reactivation and rifting.

6. Conclusions

Helium isotopes in phenocrysts from mafic volcanic rocks from south Italy (Pantelleria, Etna, Iblei, Ustica and western Aeolian islands) have an almost homogeneous $^{3}He/^{4}He$ (6.7-7.1 R_a). The different tectonic environment of these sites (stable cratonic, intraplate, subduction, rifting) appears to have no strong effect on the isotopic signature of basalts. The He-Sr-Pb isotope composition rules out an origin in deep mantle origin like FOZO and is consistent with either a HIMU younger than Cook-Austral island or with lower $^{238}U/^{204}Pb$. Helium isotopes from all Italian Plio-Quaternary volcanism correlate well with Sr, Nd and Pb isotopes. The general northward increase in radiogenic He, Sr and Pb and unradiogenic Nd reflects the progressive contamination of the mantle wedge by metasomatic fluids released by the subducting Ionian-Adriatic plate. Calculations based on the ingrowth of ^{4}He in the wedge and on the ^{4}He content of the subducting crust show that mechanisms of enrichment in radiogenic He are effective only if the wedge is He-depleted. This can be accommodated if the process of metasomatism depleted the mantle wedge.

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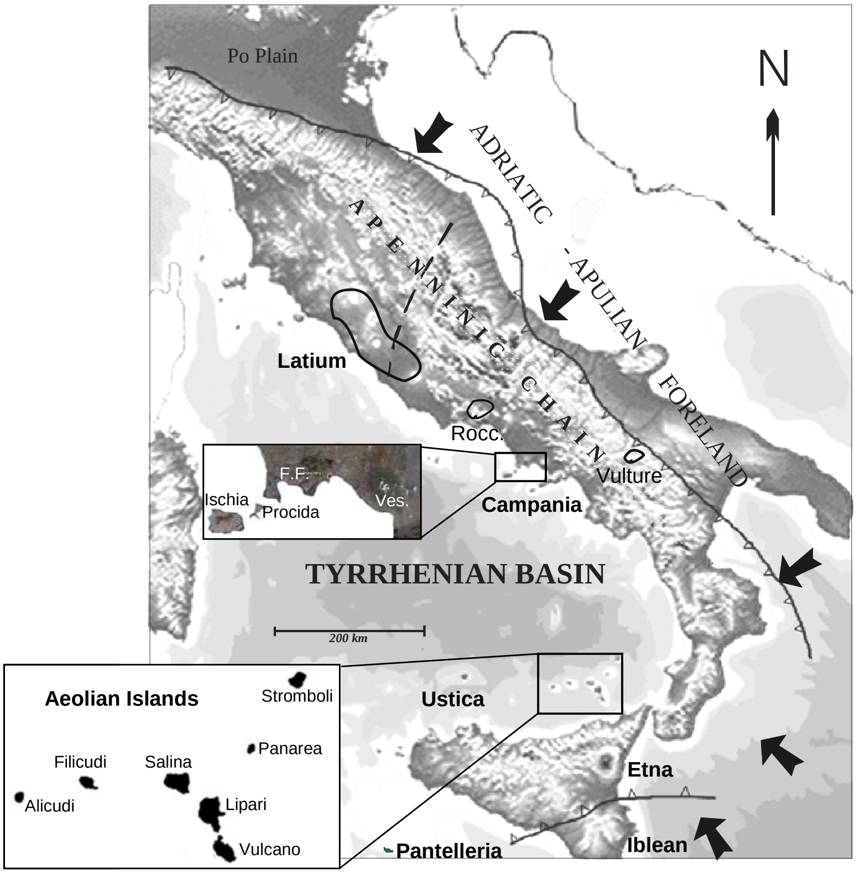


Figure 1. Map of southern Italy showing the volcanic regions sampled in this study. Material previously studied by Martelli et al. (2004) includes Latium (Alban Hills and Mt. Vulsini), Roccamonfina (Rocc.), Flegrean Fields (F.F.), Ischia, Procida and Vesuvio (Ves.). The arrows indicate the direction of the subduction. Geographic map after *Valensise and Pantosti* [2001], modified.

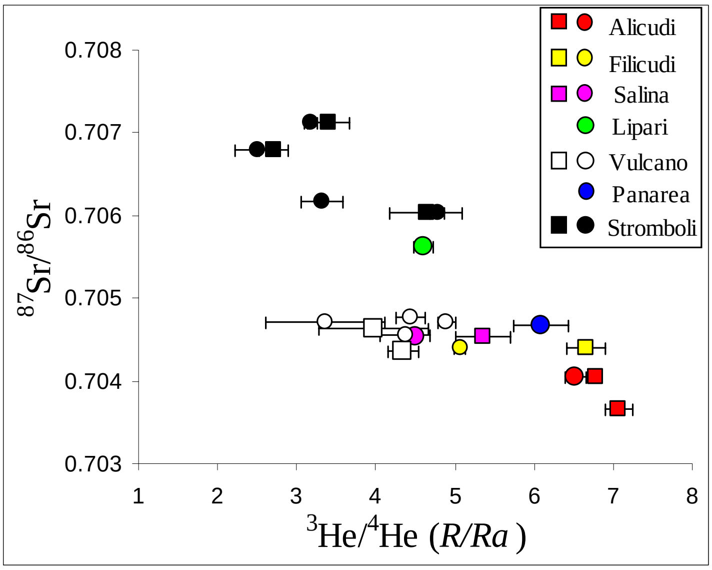


Figure 2. He-Sr isotope covariation in Plio-Quaternary basalts from Aeolian islands. Helium isotopes are determined by in vacuo crushing of olivine (squares) and pyroxene (circles) phenocrysts.

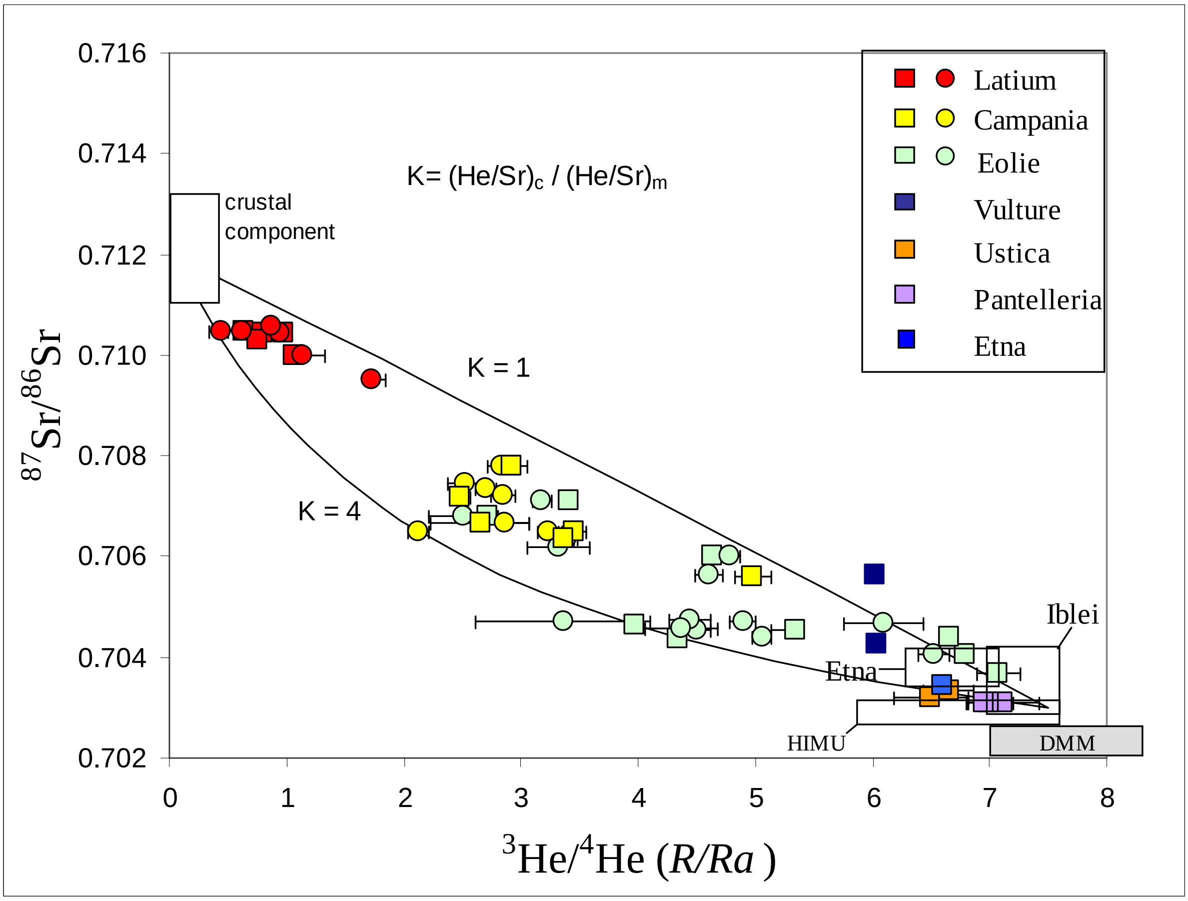


Figure 3. $^{87}Sr/^{86}Sr$ vs $^3He/^4He$ for Italian Plio-Quaternary volcanism. Helium isotopes are measured in olivine (squares) and pyroxene (circles) phenocrysts. Latium and Campania data after *Martelli et al.* [2004]. The field representing Iblei uses data from *Sapienza et al.* [2005] and *Trua et al.* [1998]. Etna data are from *Marty et al.* [1994] and this work. DMM and HIMU data after *Hofmann* [1997], *Hanyu and Kaneoka* [1997], *Farley and Neroda* [1998]. In the K notation, c: crust, m: mantle.

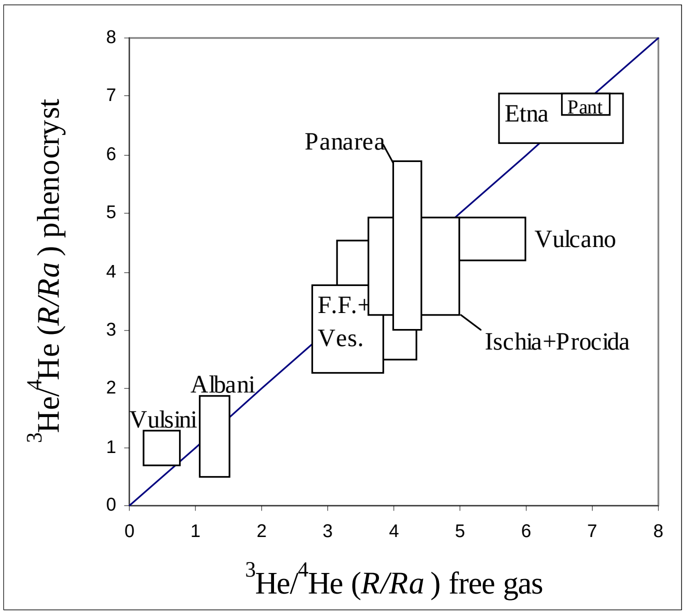
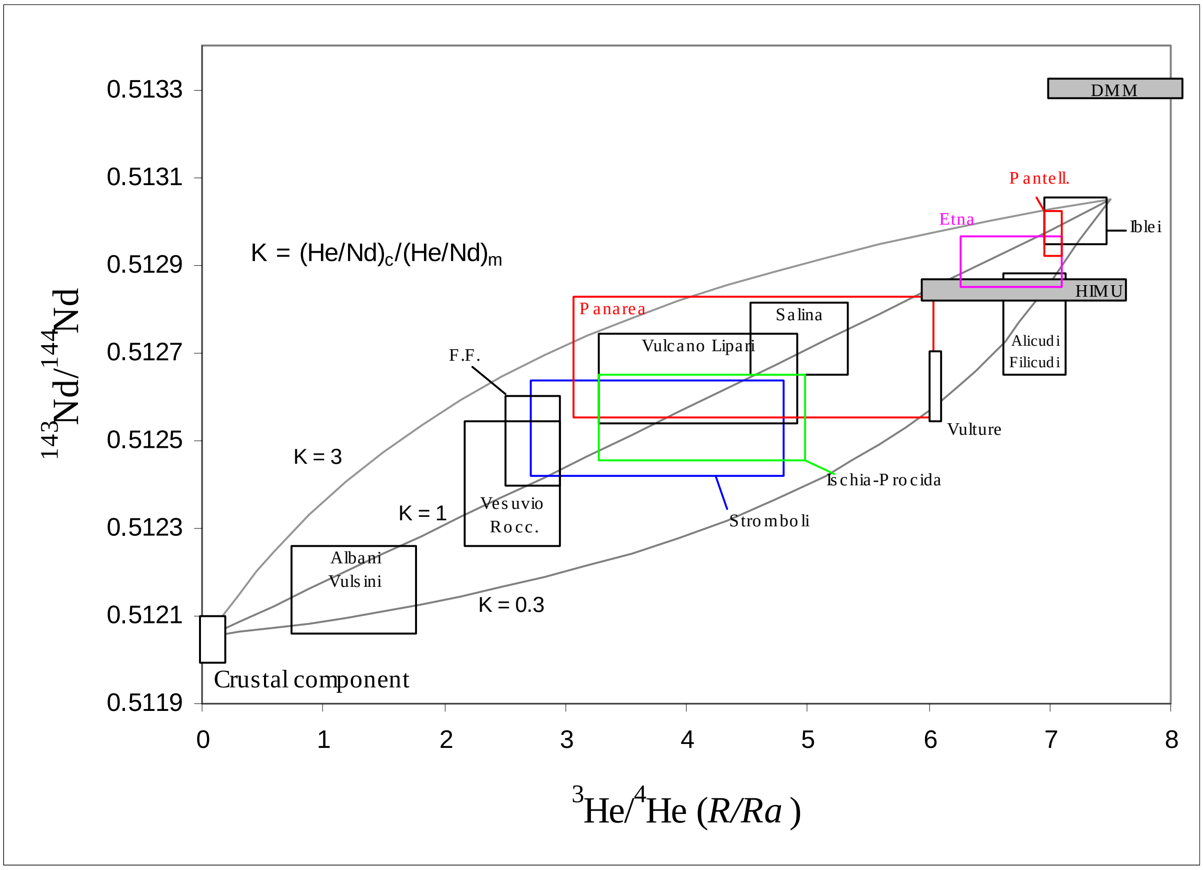


Figure 4. A comparison of the $^3He/^4He$ of phenocryst-hosted fluid inclusions with $^3He/^4He$ of free gases from the same volcanic district. Data from Vulsini, Albani, Flegrean Fields (F.F.) and Vesuvio (Ves.) are from *Martelli et al.* [2004]. Free gas data from Etna [*Caracausi et al.*, 2003], Pantelleria [*Parello et al.*, 2000], Stromboli [*Inguaggiato and Rizzo*, 2004], Vulcano [*Tedesco et al.*, 1995] and Panarea [*Caliro et al.*, 2004].


K = (He/Nd)$_c$/(He/Nd)$_m$
^{143}Nd/^{144}Nd
0.5133
0.5131
0.5129
0.5127
0.5125
0.5123
0.5121
0.5119
0
1
2
3
4
5
6
7
8
^{3}He/^{4}He (*R/Ra*)
DMM
Pantell.
Etna
Iblei
HIMU
Panarea
Salina
Vulcano Lipari
Alicudi Filicudi
F.F.
Vulture
K = 3
Ischia-Procida
Vesuvio Rocc.
Stromboli
K = 1
Albani Vulsini
K = 0.3
Crustal component

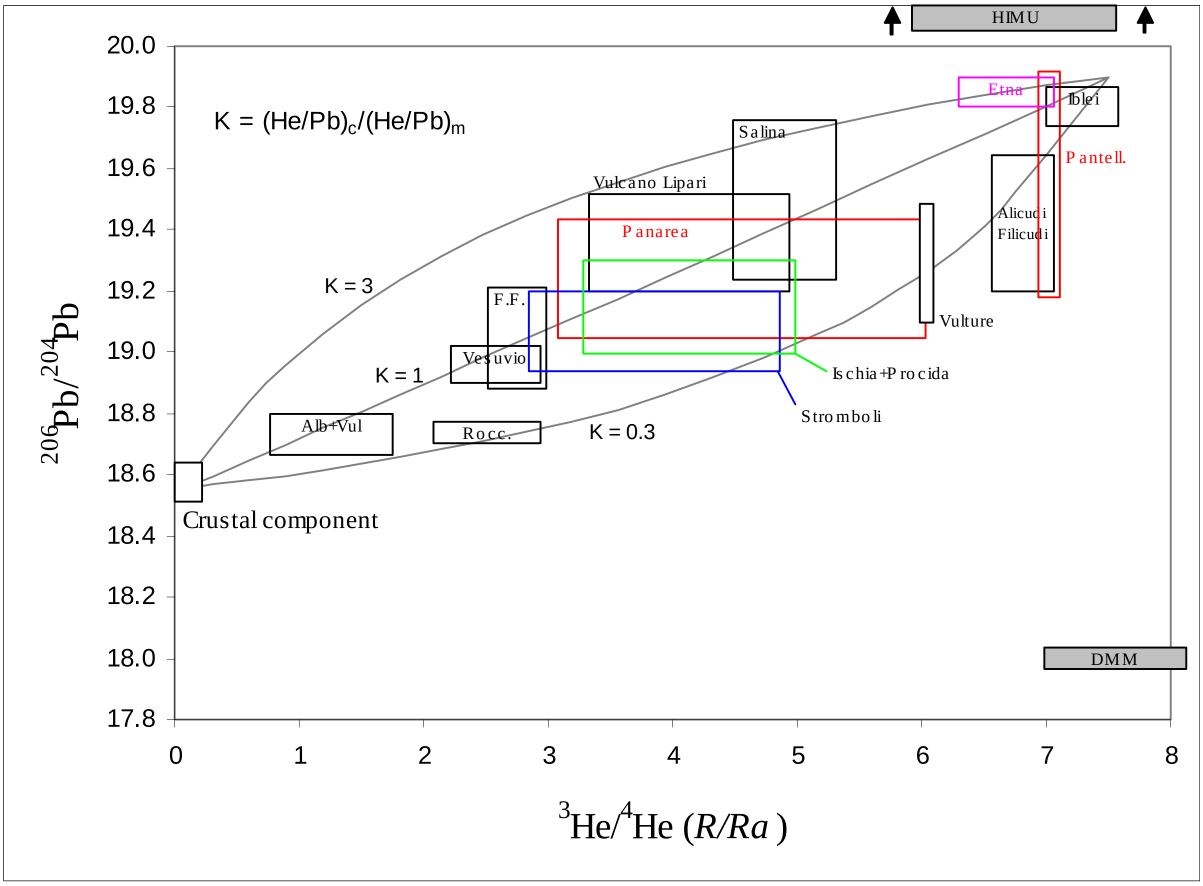

HIMU
20.0
19.8
19.6
19.4
19.2
19.0
18.8
18.6
18.4
18.2
18.0
17.8
^{206}Pb/^{204}Pb
K = (He/Pb)$_c$/(He/Pb)$_m$
Etna
Iblei
Pantell.
Salina
Vulcano Lipari
Panarea
Alicudi Filicudi
K = 3
F.F.
Vulture
Vesuvio
K = 1
Ischia+Procida
Alb+Vul
Rocc.
K = 0.3
Stromboli
Crustal component
DMM
0
1
2
3
4
5
6
7
8
^{3}He/^{4}He (*R/Ra*)

Figure 5 (a) ^{3}He/^{4}He vs. ^{143}Nd/^{144}Nd and (b) ^{3}He/^{4}He vs. ^{206}Pb/^{204}Pb constructed using our He isotope determinations [this work; *Martelli et al.*, 2004] except for Etna [*Marty et al.*, 1994] and Iblei [*Sapienza et al.*, 2005]. Pb and Nd isotope measurements after: this work; *Hawkesworth and Vollmer*, [1979]; *Ellam et al.*, [1989]; *Francalanci et al.*, [1993]; *Marty et al.*, [1994]; *D'Antonio et al.*, [1996]; *Ayuso et al.*, [1998]; *Civetta et al.*, [1998]; *Pappalardo et al.*, [1999]; *Gasperini et al.*, [2002]; *Calanchi et al.*, [2002]; *Conticelli et al.*, [2002]; *Armienti et al.*, [2004]; *Peccerillo et al.*, [2004]; *Sapienza et al.*, [2005]. DMM and HIMU data after *Hofmann* [1997]; *Hanyu and Kaneoka;* [1997]; *Farley and Neroda* [1998].

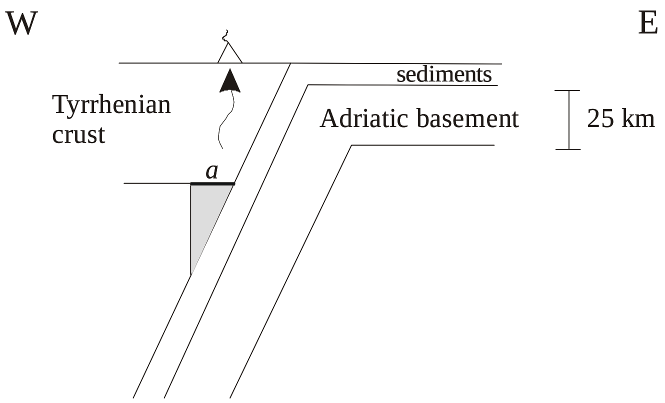


Figure 6

Simplified model used to calculate the amount of crustal He that the subducting plate may transport into the mantle wedge in the Roman Province. The structure of the Adriatic continental crust is after *Finetti et al.* [2001] and *Mele and Sandvol* [2003]. Slope of the slab is 60° [*Carminati et al.*, 2005]. The *a* segment is the E-W extension of contaminated wedge (grey area, see Figure 7).

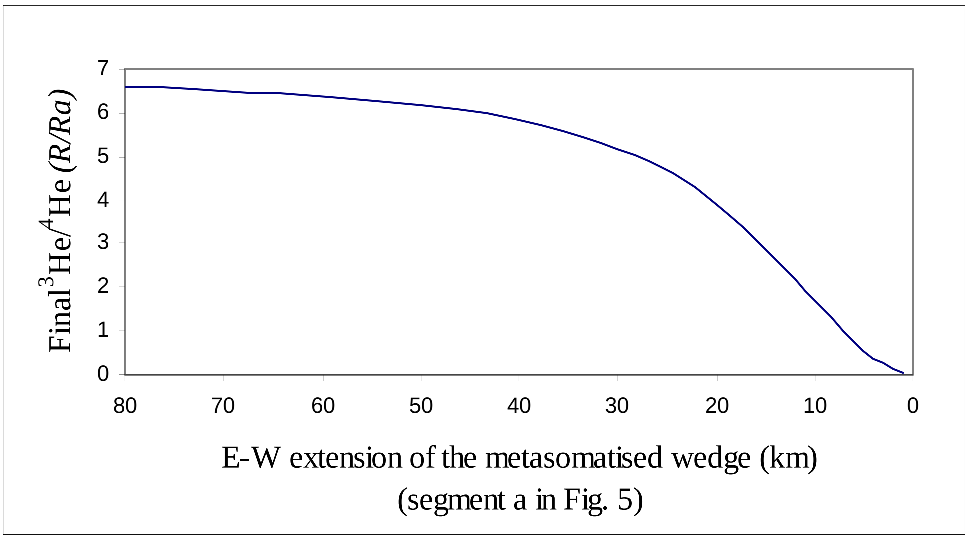


Figure 7. The effect that the direct addition of crustal radiogenic He from the subducting Adriatic plate (Table 3) will have on the ^{3}He/^{4}He of the mantle wedge beneath north Italy. Given that the slope of the slab is constrained (Figure 6), the E-W extension of the wedge allows to calculate the volume of the wedge. With the ratio $\mathrm{Volume}_{\mathrm{crust}}$ / $\mathrm{Volume}_{\mathrm{wedge}}$ we can calculate the concentration of crustal He eventually transferred into the wedge.

It is assumed that the initial mantle has ^{3}He/^{4}He = 7.6 R_a (the highest ratio measured in the area), He concentration similar to MORB-source mantle (see text) and has already been contaminated by the He produced by ingrowth. ^{3}He/^{4}He decreases significantly only if the volume of mantle wedge involved is small.

Place	**Sample**		**Location N**	**Location E**	**Weight (g)**	**^{3}He/^{4}He (R/R_a) ±1s**	**He 10^{-9} cc/g**	**^{87}Sr/^{86}Sr**
Alicudi	AL	ol	38° 33’ 13.1”	14° 21’ 42.2”	0.65	7.07 ± 0.33	2.50	0.703670 ± 15
Alicudi	GA	ol	38° 32’ 45.9”	14° 20’ 24.2”	1.19	6.79 ± 0.12	8.40	0.704051 ± 15
Alicudi	GA	px	38° 32’ 45.9”	14° 20’ 24.2”	1.40	6.52 ± 0.13	6.10	
Filicudi	Mte Guardia	ol	38° 33’ 34.4”	14° 34’ 27.1”	0.75	6.66 ± 0.24	7.00	0.704403 ± 15
Filicudi	Mte Guardia	px	38° 33’ 34.4”	14° 34’ 27.1”	1.25	5.06 ± 0.08	8.00	
Salina	T.d. Porri	ol	38° 32’ 52.8”	14° 49’ 54.0”	1.00	5.35 ± 0.34	2.30	0.704544 ± 18
Salina	T.d. Porri	px	38° 32’ 52.8”	14° 49’ 54.0”	0.91	4.50 ± 0.12	2.00	
Lipari	M.S. Angelo	px	38° 27’ 53.9”	14° 56’ 09.9”	2.30	4.60 ± 0.12	2.50	0.705629 ± 13
Vulcano	Vulcanello	ol	38° 25’ 22.6”	14° 57’ 12.4”	0.96	3.97 ± 0.69	0.33	0.704628 ± 15
Vulcano	Vulcanello	px	38° 25’ 22.6”	14° 57’ 12.4”	5.43	2.29 ± 0.40	0.096	
Vulcano	V-250	ol	38° 23’ 31.1”	14° 58’ 02.7”	0.85	4.35 ± 0.19	2.70	0.704367 ± 15
Vulcano	V-375	px	38° 23’ 50.5”	14° 59’ 10.6”	3.45	4.89 ± 0.11	6.70	0.704710 ± 15
Vulcano	Dicco	px	38° 22’ 50.0”	14° 59’ 34.8”	2.96	3.36 ± 0.75	0.19	0.704715 ± 15
Vulcano	Molineddo	px	38° 23’ 47.8”	14° 58’ 42.6”	3.68	4.44 ± 0.17	0.97	0.704761 ± 13
Vulcano	P. Luccia	px	38° 23’ 43.1”	14° 59’ 09.2”	2.00	4.37 ± 0.32	0.81	0.704560 ± 14
Panarea	P. Torrione	px	38° 37’ 46.7”	15° 04’ 16.1”	2.41	6.09 ± 0.34	1.70	0.704664 ± 15
Panarea	La Fossa	px	38° 38’ 01.0”	15° 04’ 30.0”	2.10	3.10 ± 1.75	0.042	0.705374 ± 15
Stromboli	Neo	ol	38° 48’ 33.5”	15° 13’ 30.4”	1.50	2.71 ± 0.18	4.00	0.706799 ± 13
Stromboli	Neo	px	38° 48’ 33.5”	15° 13’ 30.4”	1.53	2.51 ± 0.29	0.82	
Stromboli	2003 Biondo	ol	38° 47’ 34.8”	15° 12’ 57.7”	1.51	2.90 ± 0.37	0.022	0.706171 ± 17
Stromboli	2003 Biondo	px	38° 47’ 34.8”	15° 12’ 57.7”	0.86	3.32 ± 0.26	0.053	
Stromboli	La Petrazza	ol	38° 47’ 33.7”	15° 14’ 16.0”	0.86	4.64 ± 0.46	0.99	0.706026 ± 17
Stromboli	La Petrazza	px	38° 47’ 33.7”	15° 14’ 16.0”	2.02	4.78 ± 0.09	3.90	
Stromboli	S. Bartolo	ol	38° 48’ 23.2”	15° 14’ 10.1”	0.80	3.41 ± 0.26	1.70	0.707115 ± 16
Stromboli	S. Bartolo	px	38° 48’ 23.2”	15° 14’ 10.1”	2.21	3.18 ± 0.08	2.30	
Ustica	Faro	ol	38° 41’ 45.0”	13° 09’ 23.1”	2.02	6.65 ± 0.21	0.69	0.703322 ± 18
Ustica	Spalmatore	ol	38° 41’ 35.6”	13° 09‘ 25.2”	2.08	6.49 ± 0.32	0.28	0.703200 ± 15
Vulture*	P6	ol	40° 56’ 09.7”	15° 35’ 31.1”	6.31	6.02 ± 0.03	230	0.705648 ± 19
Vulture*	P6	px	40° 56’ 09.7”	15° 35’ 31.1”	6.13	6.06 ± 0.03	2900	
Vulture	V10	ol	40° 56’ 09.7”	15° 35’ 31.1”	6.12	6.03 ± 0.07	0.45	0.704258 ± 19
Etna	2003	ol	n.a	n.a	2.04	6.60 ± 0.10	1.40	0.703450 ± 15

Table 1. ^{3}He/^{4}He of olivine and pyroxene phenocrysts, and whole rock ^{87}Sr/^{86}Sr, of basalts from southern Italy. *Data after *Paternoster*, [2004].

Sample	**Location N**	**Location E**	**Phase**	**Weight (g)**	**^{3}He/^{4}He (*R/Ra*)**	**^{87}Sr/^{86}Sr**	**[He] (10^{-9} cc/g)**	**^{143}Nd/^{144}Nd**	**^{206}Pb/^{204}Pb**	**^{207}Pb/^{204}Pb**	**^{208}Pb/^{204}Pb**
C. Bruciata	36° 49’ 37.5	11° 57’ 22.2	ol	1.06	7.12 ± 0.30	0.70309 ± 15	1.9	0.51301 ± 1	19.637 ± 5	15.701 ± 4	39.229 ± 13
P.Guardia	36° 50’ 10.7	11° 58’ 04.3	ol	1.10	7.00 ± 0.20	0.703082 ± 15	4.2	0.51302 ± 1	19.324 ± 5	15.645 ± 4	38.913 ± 9
P.S. Leonardo	36° 50’ 09.2	11° 56’ 43.6	ol	1.15	6.95 ± 0.15	0.70311 ± 19	2.3	0.51300 ± 1	19.675 ± 5	15.672 ± 5	39.203 ± 10

Table 2. He, Sr, Nd and Pb isotopic data of Pantelleria basalt.

		REFERENCE
U in garnet	2 ppm	Extrapolated from *De Wolf et al.*, [1996]; *Jung and Mezger*, [2003]
Th/U in garnet	2	Extrapolated from *De Wolf et al.*, [1996]; *Aciego et al.*, [2003]; *Jung and Mezger*, [2003]
Garnet in the basement	1 %	Extrapolated from *Montanini and Tribuzio*, [2001]
Age of the Adriatic basement	310 Ma	*Montanini and Tribuzio*, 2001; *Finetti et al.*, [2001]
Length of subducted Adriatic crust	170 km	*Carminati et al.*, [2005]
Thickness of the basement	25 km	*Finetti et al.*, [2001]; *Carminati et al.*, [2005]
Radiogenic ^{3}He/^{4}He	0.03 *Ra*	*O'Nions and Oxburgh*, [1988]

Table 3. Parameters to calculate the amount of radiogenic He produced and accumulated in the subducting crust. We assume that He of the sediments is degassed in the early stages of the subduction while He of the basement is entirely transferred into the mantle wedge. Following *Carminati et al*. [2005], the entire basement is subducted. We considered the basement formed by the lower crustal rocks studied by *Montanini and Tribuzio* [2001]. Such parameters give an accumulation of ^{4}He of 1.1 x 10^{-6} ccSTP/g in the subducting crust.