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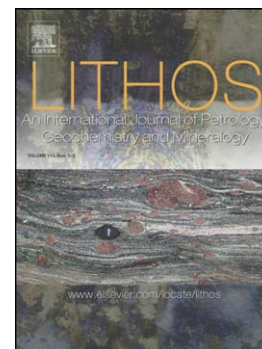
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Evidence for mantle heterogeneities in the westernmost Mediterranean from a statistical approach to volcanic petrology

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Abstract

The geological evolution of the westernmost Mediterranean region is characterised by widespread volcanic activity, with subduction (orogenic) or intraplate (anorogenic) geochemical imprints. Major, trace elements and isotopic ratios of 283 orogenic and 310 anorogenic volcanic samples from the western and central Mediterranean areas were merged in a single database that was processed using a statistical approach. Factor analysis, performed using the Principal Component Analysis (PCA) method, reduced the original 36 geochemical parameters that were expressed as oxides, elements or isotopic ratios to seven factors that account for ~84% of the variance. Combining these factors in binary diagrams clearly separates the anorogenic and orogenic fields. Anorogenic samples usually fall into a narrow compositional range, while orogenic rocks are characterised by greater variability and by alignment along different trends. These different trends are a result of large heterogeneities of the lithospheric mantle beneath the Mediterranean area because of extensive recycling of geochemically different lithologies, at least since Palaeozoic times. The results support the requirement for different mantle reservoirs in the origin of the Mediterranean volcanism. We find that the double subduction polarity model, recently proposed for the westernmost Mediterranean area, is compatible with the volcanic petrology of the last 30 My.

Keywords

Factor analysis, Geochemical database, Major elements, Trace elements, Isotopic ratios, Geodynamic evolution, Orogenic volcanism, Anorogenic volcanism.

1. Introduction

The central and western Mediterranean area encompasses the region between the Calabrian and Gibraltar arcs, including a series of Neogene marine sedimentary basins (e.g., Alboran, Valencia Trough, Algerian, Liguro-Provençal, and Tyrrhenian), the surrounding Alpine belts (e.g., Betics, Catalan Coastal Ranges, Alps, Apennines, Rif-Tell-Atlas), the Balearic Promontory, and the Corsica-Sardinia block. The whole region forms part of the western Africa-Eurasia plate boundary, where a far-field compressive stress regime was coeval with regional extension. It is the site of active and recent subduction processes, and is characterised by Neogene to Recent volcanic activity.

The central and western Mediterranean area has been widely investigated with the following aims: 1) to reconstruct the plate kinematics (e.g., Rabinowitz and LaBrecque, 1979; Dewey et al., 1989; Van der Voo, 1993; Rosenbaum et al., 2002b; Vergés and Fernández, 2012; Carminati et al., 2012; Turco et al., 2012; Casciello et al., 2015); 2) to emphasise the seismic images in order to propose snapshots of present-day lithospheric domains beneath Europe and the Mediterranean (e.g., Gutscher et al., 2002; Piromallo and Morelli, 2003; Spakman and Wortel, 2004; Wortel et al., 2009; Giacomuzzi et al., 2012; Bezada et al., 2013; Palomeras et al., 2014; Thurner et al., 2014; Chiarabba et al., 2014); and 3) to describe the volcanic activity that affected several areas from Spain to northern Italy and from northern Africa to central Europe since the Oligocene (El Azzouzi et al., 1999; Coulon et al., 2002; Beccaluva et al., 2004, 2013; Harangi et al., 2006; Lustrino and Wilson, 2007; Lustrino et al., 2011; Carminati et al., 2012; El Azzouzi et al., 2014). The volcanic activity in the central and western Mediterranean region can be broadly divided into two main types (Fig. 1) on the basis of their tectonic and geochemical features (see Lustrino and Wilson, 2007 and Lustrino et al., 2011 for reviews). Orogenic or subduction-related volcanism usually refers to volcanic rocks generated by continent-continent collision or are associated with subduction of oceanic

lithosphere (e.g., Carminati and Doglioni, 2004). It comprises arc tholeiitic, calc-alkaline, high potassium (HK) calc-alkaline, shoshonitic and ultrapotassic products. Anorogenic or intraplate volcanism is associated with a post-collisional environment and includes alkaline rocks with sodic affinity. Combining this information, several and sometimes competing geodynamic models have been put forward to explain the Tertiary tectonic evolution of the western Mediterranean area (Doblas et al., 2007).

Tomographic models have resulted in a consensus on the extent, shape and dip of the present-day subducting slabs in the region. However, discrepancies arise concerning the initiation of subduction relative to the extent of the subduction front, its orientation and dip. Some authors have proposed an initial N-NW dipping subduction front that extended from the Alps to the present-day Gibraltar Arc (e.g., Faccenna et al., 2004; Schettino and Turco, 2006; Carminati et al., 2012). Other authors proposed restrictions on the initial N-NW dipping subduction zone from the Alps to the southwest termination of the present-day Balearic Promontory (e.g., Rosenbaum and Lister, 2004; Spakman and Wortel, 2004; van Hinsbergen et al., 2014). A third model proposed a lateral change in the initial subduction polarity from S-SE along the north Moroccan margin to N-NW along the Balearic Promontory to the Alps (Vergés and Fernández, 2012; Casciello et al., 2015).

The incorporation of volcanism into these models is uneven. Duggen et al. (2004, 2005) explained the geochemical features of westernmost Mediterranean volcanism as a result of delamination in a context of a westward roll-back phase. Booth-Rea et al. (2007) used the volcanism to support a model in which E-dipping subduction beneath the Alboran and Algero-Balearic basins was active until the late Miocene, when it stopped, probably because of the complete consumption of the oceanic crust. The Si-K-rich volcanism is interpreted as a result of anatexis because of the direct contact of the

asthenosphere with the base of the crust, driven by delamination of the lithospheric mantle (Bird, 1979). Wilson and Bianchini (1999) provided clues for the origin of the most primitive mafic igneous rocks that can be considered as the best indicator for the initial geodynamic setting. Harangi et al. (2006) dealt with the subduction-related volcanic rocks and their relation with competing geodynamic models proposed for the evolution of the central and western Mediterranean. Finally, Lustrino and different co-workers (Lustrino and Wilson, 2007; Lustrino et al., 2011; Lustrino, 2011) presented an overview of the two main geochemically different magma-types (anorogenic and orogenic), with an aim of providing a general model for the petrogenesis of the intraplate affinity suite and a possible relationship between tectonic evolution and geochemistry of the subduction-related rocks.

The huge amount of available information on the geochemistry of the volcanic products in the western and central Mediterranean area makes its incorporation into the proposed geodynamic models difficult, mainly due to the extreme heterogeneity of the data. A way to manage large numbers of variables with no obvious relationships among them is to use a factor analysis technique. Although this technique is used widely in the social and life sciences and is becoming a common method in earth science, it is not yet used in volcanic geochemistry. In this paper, we present the results obtained by applying a factor analysis (using the principal component method) to a large database composed of Mediterranean orogenic (Lustrino et al., 2011) and anorogenic (Lustrino and Wilson, 2007) volcanic rocks. Factor analysis is here used to remove redundant variables and to replace the entire geochemical data file by a smaller number of variables in order to support previous geochemical interpretations and to ascertain the utility of the method.

Our main aims are: 1) to verify whether a factor analysis technique can be applied to geochemical issues; 2) to highlight the differences in geochemical composition of the

intraplate affinity and subduction-related volcanic rocks of the western and central Mediterranean, and the evolution of these differences over time and 3) to constrain the origin of the mantle heterogeneities which are present in the source of the volcanism. Furthermore, we analyse the compatibility between the geochemical signature of the volcanism in the westernmost Mediterranean and a recently proposed geodynamic model based on a lateral change of subduction polarity (Vergés and Fernàndez, 2012; Casciello et al., 2015).

2. Cenozoic tectonic evolution of the Central and Western Mediterranean

The central and western Mediterranean region has undergone a complex geodynamic evolution since the early Cretaceous. This complex history is recorded in the Alpine orogeny (Dal Piaz et al., 2003), in the numerous areas where magmatism and orogenesis occurred (Araña et al., 1983; Cebriá and López-Ruiz, 1995; Harangi et al., 2006; Lustrino and Wilson, 2007; Lustrino et al., 2011; Beccaluva et al., 2011, 2013), and in the formation of several back-arc basins that opened because of roll-back of lithospheric slabs with various retreat directions (e.g., Carminati et al., 2012). These basins (Fig. 1) are the Liguro-Provençal and the Tyrrhenian basins (Van der Voo, 1993; Faccenna et al., 1997; Doglioni et al., 2004), the Valencia Trough and the Algerian Basin (Martí et al., 1992; Torne, 1996; Vergés and Sàbat, 1999; Ayala et al., 2003; Roca et al., 2004; Carballo et al., 2015), and the Alboran Basin (Comas et al., 1992, 1999; García-Dueñas et al., 1992; Torne et al., 2000; Frizon de Lamotte et al., 2004; Soto et al., 2008). The overall tectonic framework in which this evolution occurred is characterised by convergence between the African and Eurasian plates caused by the opening of the

southern Atlantic Ocean. From Early Cretaceous times on, Africa moved slowly toward Eurasia with a NE-directed counter-clockwise rotation (Dewey et al., 1989; Mazzoli and Helman, 1994; Rosenbaum et al., 2002a), shifting to a NNW direction at about 40 Ma (Rosenbaum et al., 2002a; 2002b), and finally to a N-S direction (Devoti et al., 2008).

It is widely accepted that the evolution of the central Mediterranean area is linked to the roll-back of Tethyan oceanic lithosphere that had remained landlocked since the Oligocene. However, despite the consensus among mantle tomography models that image a subducted and partly torn slab beneath the Betic-Rif orogen (e.g., Spakman and Wortel, 2004; Díaz et al., 2012; Miller et al., 2013; Bonnín et al., 2014; Thurner et al., 2014; Fichtner and Villaseñor, 2015), the debate focuses on how to trace the evolution of these subducted plates back to their initial configuration.

The earliest tectonic reconstructions considered that the central and western Mediterranean basins and surrounding chains resulted from extensional stresses that dismantled the western Alpine belt. This orogenic dismembering produced the radial dispersion of its high pressure–low temperature (HP-LT) ‘collisional’ metamorphic rocks to form the Calabria-Peloritani, Kabilyes and Alboran terranes (Alvarez et al., 1974; Boccaletti and Guazzone, 1974; Alvarez and Shimabukuro, 2009). As argued in Casciello et al. (2015), this initial interpretation had a strong influence on later reconstructions that, in the case of the westernmost Mediterranean area, can be grouped into three different hypotheses:

- 1) The original Alpine belt formed a continuous chain from the present-day Alps to the Gibraltar area and was related to NW-directed subduction, at least since the middle Eocene, incorporating the HP-LT metamorphic domains of the future Betic-Rif Internal Zone (Alboran Domain), the Kabylies blocks, and the Calabria-Peloritani units. Further

retreat and slab roll-back towards the SE caused dispersal of the metamorphic units following a radial pattern. The Calabria-Peloritani units moved ESE together with the Sardinia-Corsica block during the opening of the Liguro-Provençal Basin, whereas the Kabylies shifted southwards during the opening of the Algerian Basin (e.g., Jolivet and Faccenna, 2000; Faccenna et al., 2004; Faccenna et al., 2014). These authors introduced the concept of slab tears in the subducting lithosphere to allow for an independent WSW roll-back in the Gibraltar area, with respect to the overall S-directed roll-back of the Algerian slab. The main criticism of this model is that the formation of the Miocene NW-verging Betic orogen and the S- to E-dipping present-day subduction polarity are poorly explained.

2) A second model proposes that the initial N-NW dipping subduction zone extended from the Alps to the south of the present position of the Balearic Promontory (e.g., Lonergan and White, 1997; Rosenbaum and Lister, 2004; Spakman and Wortel, 2004; van Hinsbergen et al., 2014). Similar to the previous model, this orogenic belt was composed of HP-LT metamorphic units that dismembered radially, because of slab roll-back. The main difference with the previous model is that the segment of the subduction front, associated with the present Balearic block, underwent a clockwise rotation of $\sim 180^\circ$ which was accommodated by lithospheric tearing along the North African margin. This allowed for over 500 km of westward movement of the Alboran domain and the E-W opening of the Algerian basin (van Hinsbergen et al., 2014).

3) The latest interpretation has been proposed by Vergés and Fernàndez (2012). Based on the position of high-grade metamorphic rocks with respect to subduction polarity and several other geological and geophysical constraints, these authors suggested that the Betic-Rif Arc originated from the Oligocene-Miocene north-westward and westward roll-back of the southeast-dipping subduction of Ligurian Tethys lithosphere below

Africa. In this view, the Late Oligocene-Miocene evolution of the central and western Mediterranean is linked to two opposite dipping subductions: a northwest-dipping, southeast-retreating subduction related to the migration of the Kabylies units, and a southeast-dipping, northwest-retreating subduction related to the formation of the Betic-Rif orogen.

3. Geochemical data and factor analysis

Two large databases in the literature include much of the volcanic activity that has occurred in the Mediterranean area from the Oligocene to the present-day (Fig. 1). A compilation of the orogenic-related rocks has been published by Harangi et al. (2006) and more recently by Lustrino et al. (2011), while the geochemical data of the anorogenic rocks were gathered by Lustrino and Wilson (2007), and further updated by Lustrino (2011). For comparison, we also considered the Canary Islands dataset that, as expected, shows a monotonous and unambiguous intraplate signature from Oligocene to Recent.

The main limitation of such large datasets is visualisation of the wealth of information they contain. In geochemistry, binary or ternary diagrams are used to identify significant variation in chemistry, combining major, trace elements and isotopic ratios. However, even if elemental ratios are considered, it is highly unlikely to observe, at the same time, variations of more than six elements. Hence, typically, several diagrams are used to display the chemical data, which often distract the attention of the researcher from the final goal of unravelling the geochemical evolution of a suite of rocks, and to insert them in a coherent geodynamic framework. In this respect, a statistical approach

through factor analysis can help, as a powerful tool to combine more elements and allowing us to summarise geochemical variations in a few binary diagrams.

The orogenic volcanic dataset of Lustrino et al. (2011) is a compilation of >7,000 samples, while its anorogenic counterpart accounts for ~7,800 analyses (Lustrino, 2011). These databases have been merged and filtered by choosing those samples that are characterised by the most complete geochemical dataset in terms of:

- a) Major elements, expressed as oxides of Si, Ti, Al, Fe, Mn, Mg, Ca, Na, K, P.
- b) Trace elements, including Large Ion Lithophile Elements (LILE) - Cs, Rb, Ba, Sr and Pb; High Field Strength Elements (HFSE) - Th, U, Nb, Ta, Zr and Hf; and Rare Earth Elements (REE) - La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Y and Lu.
- c) Isotopic composition, mainly expressed by $^{87}\text{Sr}/^{86}\text{Sr}$, $^{143}\text{Nd}/^{144}\text{Nd}$, $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$, $^{208}\text{Pb}/^{204}\text{Pb}$, $^{208}\text{Pb}/^{206}\text{Pb}$ and $^{207}\text{Pb}/^{206}\text{Pb}$.

After filtering, a total of 283 orogenic and 310 anorogenic volcanic samples were selected (Fig. 2). This procedure led us to completely exclude samples from Algeria and Tunisia, and to select only three samples from Sardinia and one from Corsica (Sisco). We built a 593 x 43 matrix in which each row represents a sample and each column represents a variable (i.e., a geochemical element characterising the sample). This matrix was processed in order to obtain standardised values that are expressed as $Z_i = (x_i - x_m)/\sigma$, where x_i is the non-standardised value of variable x , x_m is the mean value of variable x , and σ is the standard deviation of variable x . The standardisation procedure inflates variables whose variance is small, reduces the influence of variables whose variance is large, and removes the influence of different measurement units in the data by making them dimensionless.

Factor analysis is a multivariate statistical method used to represent a large number of variables in the original data by a substantially smaller number of factors. The term factor in geochemistry refers to controlling processes and factor analysis is ideally suited for an easy presentation of the essential information in a geochemical dataset (Reimann et al., 2002). The aim of statistical analysis is to reduce the dimensionality of the original space, thus allowing for a more comprehensible interpretation of a reduced number of dimensions which underlie the old dimensions (Rietveld and Van Hout, 1993).

For this purpose, the matrix has been processed using the Statistical Package for Social Sciences (SPSS) version 17, which allowed a first reduction of the number of variables to describe the 593 samples by reading the communality value (between 0 and 1). The communality of a variable is an automatically calculated parameter, based on the sum of the loadings of this variable on all of the extracted factors, which reflects the percentage of variance in a given variable that jointly encompasses all of the factors (interpreted as the reliability of the indicator). Those variables with communality <0.6 were considered meaningless and were disregarded. The matrix was reprocessed until all variables had communality >0.6 , thus excluding MnO, Cs, Ta, Hf, Gd, Er and $^{207}\text{Pb}/^{204}\text{Pb}$ isotopic ratio in the final factor analysis.

The starting point of factor analysis is a correlation matrix obtained by calculating the correlations between each pair of variables. Extraction of the factors is undertaken by calculating the eigenvalues of the correlation matrix. The number of positive eigenvalues determines the number of dimensions needed to describe the set of scores without losing information, and the number of extracted factors (Rietveld and Van Hout, 1993). Construction of the factor is based on a transformation matrix that is given by the eigenvectors of the eigenvalues (Rietveld and Van Hout, 1993). Once the factors

have been extracted, the factor loadings are calculated by correlating the original variables and the newly obtained factors. The first factor accounts for the maximum part of the variance, the second factor accounts for the maximum part of the remaining variance, and so on. In our case, seven factors (F) were extracted that account for ~84% of the cumulative variance (F1 22.7%; F2 18.6%; F3 12.2%; F4 11.8%; F5 6.4%; F6 6.4%; F7 6.1%) (**Table 1**).

The interpretation of the obtained factors can be problematic because the original variables have high loadings on the first factors as they account for most of the variance. To get around this problem, an orthogonal (Varimax) factor rotation is applied. Finally, each standardised variable score is multiplied by the corresponding standardised scoring coefficient, and the sum of these products results in the factor scores (Supplementary Material). In our case, the number of orogenic and anorogenic samples must be similar in order to avoid the dominance of one over another in extracting the main factors.

The factor scores are plotted in binary diagrams (Supplementary Material) in order to observe the variations affecting the samples in terms of controlling processes. Considering that the factors are combinations of all the variables used for the geochemical description and that each factor is standardised to a mean value of 0 and a variance of 1, the intersection of the axes represents the mean of all the variables. For example, in the F2 vs. F1, diagram the intersection of the axes represents the point where the values of the variables, that are weighted by these two factors, coincide with their mean. Moving away from the axes intersection, the values of the variables will increase (positive side of the axes) or decrease (negative side of the axes) with respect to the mean.

4. Outcomes of the factor analysis

F1 is characterised by a positive correlation with K_2O , $^{87}Sr/^{86}Sr$, Rb, Sm and a group of highly mobile elements such as U, Th and Pb, and by a negative correlation with Fe_2O_3 , CaO and $^{143}Nd/^{144}Nd$ (**Table 2**). F2 depends positively on Sr and Ba, as well as on some Light REE (LREE; La, Ce and Nd), Medium REE (MREE; Eu, Dy) and Y. F3 shows a positive correlation with TiO_2 , MgO and P_2O_5 and a negative correlation with SiO_2 and Al_2O_3 . F4 depends positively on Heavy REE (HREE) and Pr (LREE), while F5 is dependent on Na_2O , Zr and Nb. Statistically, Pr is correlated with F4, although most of the LREE are associated with F2. Finally F6 and F7 depend positively on Pb isotopic ratios. F6 is correlated with $^{208}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$, whereas F7 correlates to $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$. By combining the seven factors in binary diagrams (Supplementary Material), we observed that factors F1, F2, F3 and F5 explain the majority of the chemical variations. Fig. 3 shows the plots of the rotated component matrix expressed as binary diagrams of F2 vs. F1, F3 vs. F1 and F5 vs. F3. These plots help in understanding what variables influence the different factors more. On the other hand, in Fig. 4 and Fig. 5 we use the factor scores obtained for each sample in order to understand how the volcanic rocks in this study are related to the different factors (i.e., to the different original variable or chemical components).

On the whole, both orogenic and anorogenic samples show wide variations of all the factors (Fig. 4 and Supplementary Material). Most localities show a wide compositional range, in terms of both F1 and F2 (Fig. 5A and Fig. 5B), except Sardinia and Corsica (Sisco) which have very few samples. Orogenic samples from Campania and the Aeolian Islands mostly straddle the zero axis intersection (Fig. 5A), indicating that their compositions are close to the mean values of all variables included in F1 and F2. No

correlation is found between composition and age, except some recent Campanian volcanic products shifting towards higher F1 values that are characterised by an increase in K₂O, Rb, Sm, U, Th and Pb contents, as well as radiogenic ⁸⁷Sr/⁸⁶Sr. On the other hand, samples of the Roman Province, Western Alps and Tuscany, mostly plotting on the positive side of the x-axis, show alignments along different trends (Fig. 5A). Note that Tuscany samples include some rocks from Capraia but not from Elba Island. Three features can be highlighted: 1) Roman Province volcanic products show a very wide compositional range in terms of both F1 and F2, thus being characterised by an increase of K₂O, Rb, Sm, U, Th, Pb, and more radiogenic ⁸⁷Sr/⁸⁶Sr, coupled with a decrease of Fe₂O₃ and CaO, and less radiogenic ¹⁴³Nd/¹⁴⁴Nd, as well as by enrichment in Sr, Ba, LREE, MREE and Y contents; 2) All samples from Tuscany plot on the negative side of the F2-axis, highlighting a lower content of Sr, Ba, LREE, MREE and Y with respect to the Roman Province volcanics. One of the Tuscany lamproites shows a lower LREE content than the Roman province samples. At the same time, Tuscany samples show an increase in elements related to F1, with the highest contents observed in lamproites. Western Alps samples plot between the Roman and Tuscan provinces trends; 3) There is a temporal correlation of the volcanic products of Tuscany, with the lamproites (4.1 Ma) associated with calc-alkaline, HK calc-alkaline and shoshonitic rocks (0.88-1.35 Ma), being the first in the eruption sequence (Ferrara and Tonarini, 1985; Conticelli et al., 2004; and references therein).

In the westernmost Mediterranean (Fig. 5B), the orogenic samples of southern Spain, Morocco and Alboran Basin (Fig. 1) are mostly aligned along a unique trend that starts from slightly negative F1 and F2 values and evolves towards higher F1 and slightly lower F2 values. Lamproites of southern Spain clearly plot outside this trend, between -1 and +1 F2 values, and are characterised by the highest F1 values, indicating highest

K₂O, Rb, Sm, U, Th and Pb contents and the highest ⁸⁷Sr/⁸⁶Sr, coupled with the lowest Fe₂O₃ and CaO contents and the lowest ¹⁴³Nd/¹⁴⁴Nd. In contrast to the Tuscany rocks, the Spanish volcanic rocks (lamproites, calc-alkaline, HK calc-alkaline and shoshonitic rocks) are characterised by an opposite time sequence (Conticelli et al., 2009a). Lamproitic volcanism started later (8.08 - 6.37 Ma) than the other subduction-related volcanic activity (14.4 - 6.2 Ma; Turner et al., 1999; Duggen et al., 2004, 2005; and references therein).

Both central and western Mediterranean anorogenic rocks show the same F1 and F2 variations (light blue fields in Fig. 5A and Fig. 5B). The widest compositional range is observed in the Upper Cretaceous rocks of Portugal; all the others fall on the negative side of F1. The major variation for these rocks is observed for Sr, Ba, LREE, MREE and Y contents (i.e., F2) and no age correlation is highlighted.

Fig. 3B, and consequently the values of the factor scores of Fig. 5C and Fig. 5D, give a unique opportunity to observe the variation of all major elements at the same time (except MnO, which is excluded from the factor analysis, and Na₂O, which is included in F5). Subduction-related volcanics mostly plot on the negative side of the y-axis, i.e., they are usually characterised by low TiO₂, MgO, P₂O₅ and high SiO₂ and Al₂O₃ contents. Lamproites of both central and western Mediterranean areas show high values of F1 and F3, unlike all the other volcanic rocks. The samples from the central and western Mediterranean with intraplate affinity are characterised by a wide F3 variation, with a group of rocks showing the highest SiO₂ and Al₂O₃, and lowest TiO₂, MgO, P₂O₅ contents, mostly belonging to Palaeocene German and upper Cretaceous Portuguese suites.

A general tendency observed in Fig. 5E and Fig. 5F is that, as expected, the orogenic samples are mostly characterised by negative values of F5, i.e., lower Na₂O, Zr and Nb contents than the anorogenic rocks, which generally plot above the zero axes with Na₂O/K₂O ~1. Furthermore, subduction-related rocks of both central and western Mediterranean are aligned along a trend with factors F5 and F3 being inversely correlated. Lamproitic rocks are very well discriminated compared to other samples (light red field in Fig. 5E and Fig. 5F) and are characterised by a positive correlation of factors F3 and F5, such that an increase of Na₂O, Zr and Nb is associated with an increase in TiO₂, MgO and P₂O₅, and a decrease of SiO₂ and Al₂O₃.

5. Mantle heterogeneities beneath the Mediterranean region

Volcanic rocks are characterised by geochemical signatures that are typical of the environment in which they are produced. However, processes other than simple partial melting have to be accounted for in order to explain their geochemistry. These processes can be related to metasomatic and refertilisation events in the mantle. Recycling of material via subduction processes can also modify the mantle composition, resulting in variations of the erupted volcanic products. In the central and western Mediterranean, several recycling events of various continental/oceanic subducted domains could have contaminated the lithospheric mantle for millions of years (Fig. 6). These episodes could be as old as Triassic and Permian periods, and be related to the Variscan Orogeny and the subsequent Pangea Triassic rifting processes that pre-date the central Atlantic opening. Most of the geodynamic models that have been proposed for the central and westernmost Mediterranean (Tyrrhenian area, the Alps, southern Spain, and northern Morocco) invoke upwelling of sublithospheric mantle. This upwelling would be

responsible for the erupted anorogenic suites, together with subduction-related rocks (Turner et al., 1999; Lustrino et al., 2004; Duggen et al., 2005; Harangi et al., 2006) (Fig. 1).

The applied statistical approach permits identification of the major geochemical components through the observation of a considerably smaller number of binary diagrams, obtained by combining major, trace elements and isotopic ratios. In Fig. 4 and Fig. 5, orogenic and anorogenic rocks are generally well separated with some overlapping regions, the orogenic samples being characterised by a greater compositional variability that highlights the heterogeneity of their mantle source. When combining F1 and F2, or F1 and F3, various evolutionary trends can be observed that affect the Roman Province, Tuscany or the Western Alps. These trends reflect recycling of material of different origins that was introduced via subduction and caused metasomatism of the pristine mantle that eventually melted. The two end-members represented by the Tuscan and Roman provinces are clearly related to different types of material brought down by the subducting slab, which resulted in different petrographic and geochemical features of the erupted magmas. In the Roman Province, slab-derived melts have metasomatised a fertile mantle, while beneath Tuscany a harzburgitic mantle could have been metasomatised through the subduction of continental crust or sediments (Avanzinelli et al., 2009). Both provinces are characterised by an evolution of their volcanism towards potassic and ultrapotassic end-members. In the Roman Province, the volcanic products evolved from calc-alkaline to HK calc-alkaline and shoshonitic SiO_2 -saturated (leucite-free), to strongly SiO_2 -undersaturated leucite tephrites and phonolites (Peccerillo, 2005; Avanzinelli et al., 2009; Conticelli et al., 2009b). The volcanic products from Tuscany evolved to ultrapotassic leucite-free

lamproites, characterised by relatively high MgO and SiO₂ with crustal isotopic signature (Conticelli et al., 2009a, and references therein).

The benefits of the statistical approach in geochemical investigations are provided by the discrimination of the orogenic and anorogenic fields, as well as that of the lamproitic rocks. Other evidence in favour of the proposed approach is provided by the combination of the elements within each factor. The positive correlation between K₂O and Rb or U-Th-Pb, as well as the negative correlation between the ⁸⁷Sr/⁸⁶Sr and ¹⁴³Nd/¹⁴⁴Nd isotopic ratios on F1, are clear examples. The negative correlation between MgO and SiO₂-Al₂O₃ recorded also by F3, or the dependence of F4 almost entirely on HREE, further confirm the appropriateness of the approach. Supporting evidence is, finally, also given by F6 and F7 depending on ²⁰⁸Pb/²⁰⁶Pb - ²⁰⁷Pb/²⁰⁶Pb, and ²⁰⁶Pb/²⁰⁴Pb - ²⁰⁸Pb/²⁰⁴Pb, respectively.

6. Volcanic activity and the double polarity subduction model

Geodynamic models that have been proposed for the evolution of the westernmost Mediterranean are mainly based on geologic and seismologic evidence, as well as on petrography and exhumation ages of metamorphic units that compose the inner parts of the Betic-Rif arc. Many of these models are poorly constrained from a volcanological point of view, and volcanic activity is not only used as a key constraint but also as a corollary. The complexity in interpreting the volcanic geochemistry in this region is basically related to the long-term recycling of material, coupled to depletion events caused by partial melting due to subduction or rifting phases (Fig. 6). Modifications of the Mediterranean mantle are, therefore, reflected in the volcanic products, whose

geochemical features do not always correspond to the present-day geodynamic environment in which they are sampled (see also Lustrino and Wilson, 2007; Lustrino et al., 2011; Carminati et al., 2012).

Here we analyse the geochemical characteristics of the volcanic rocks in the framework of the recent geodynamic model proposed by Vergés and Fernández (2012) for the westernmost Mediterranean area. These authors proposed the existence of two opposite dipping slabs (Fig. 6), which retreated in opposite directions (Fig. 7), based on four key premises related to: 1) the large-scale plate reconstructions addressing the Africa–Iberia plate boundary during the Mesozoic and Cenozoic; 2) the geographic distribution of HP/LT metamorphic rocks of the inner orogeny located along the Alboran and Algerian margins; 3) the role of mechanical stratigraphy of the Iberia paleo-margin, where the External Units are separated from the Internal Betic–Rif units through Triassic detachment levels; and 4) the timing of onset of compressional deformation along the Betic–Rif system.

This model includes seven time-steps or stages. The first stage was related to the onset of Africa-Iberia convergence at about 85 Ma, and volcanic activity was restricted to southwest Iberia far from the Ligurian-Tethys domain. The second stage (middle Eocene, 47–42 Ma) takes into account a HP/LT metamorphic peak (recorded in the Nevado-Filabride complex) related to SE subduction beneath northern Africa. During this stage, no volcanic activity occurred in the westernmost Mediterranean, but some anorogenic volcanic rocks are found in the High Atlas that are probably unrelated to subduction (at Tamazert and Rekkam; Bouabdli et al., 1988; Bernard-Griffiths et al., 1991; Bouabdli and Liotard, 1992; Rachdi et al., 1997; Duggen et al., 2005).

In the third stage (Oligocene, 30-25 Ma), the roll-back phase and exhumation of the HP/LT Ligurian-Tethys metamorphic rocks began. The earliest magmatic manifestation was intrusion of tholeiitic Malaga dykes, dated at 33.6 ± 0.6 Ma (Duggen et al., 2004) and 30.2 ± 0.9 Ma (Turner et al., 1999). Younger ages ranging from 23 to 17 Ma have been reported by several authors (e.g., Torres-Roldán et al., 1986; Turner et al., 1999; Duggen et al., 2004), coinciding with metamorphic events dated at 22-17 Ma (Priem et al., 1979; Monié et al., 1994; Kelley and Platt, 1999). These young volcanic ages (23-17 Ma) are related to a thermal event that reset the system, rather than being related to magma ascent and dyke emplacement (Duggen et al., 2004; Platt et al., 2013). The tholeiitic Malaga dykes are characterised by moderate Fe-enrichment (Turner et al., 1999). They show crustal contamination, indicated by small negative Ta-Nb, Ti and Zr-Hf anomalies, positive Rb, U, K and Pb anomalies, higher $^{87}\text{Sr}/^{86}\text{Sr}$, and lower $^{143}\text{Nd}/^{144}\text{Nd}$ relative to the depleted mantle, and elevated $^{208}\text{Pb}/^{204}\text{Pb}$ and $^{207}\text{Pb}/^{204}\text{Pb}$. However, the least contaminated samples show relatively low Fe and high Si contents, flat REE patterns and high HREE concentrations, indicating that partial melting did not take place in the presence of garnet. Turner et al. (1999) calculated 10-15% of mantle partial melting in the spinel stability field (less than 60-70 km). This allows us to place the genesis of these melts as the results of partial melting of a mantle wedge (Duggen et al., 2004), most probably enriched in incompatible elements through percolation of fluids from the subducting slab and involving a sedimentary component, in agreement with the third stage, described by Vergés and Fernández (2012). The Malaguide Complex corresponds to the upper plate and has been tectonically transported to its present position as a consequence of the slab roll-back.

The fourth stage, dated as late Oligocene-early Miocene (27-20 Ma), was characterised by a progressive northwest and westward displacement of the trench resulting in an

arcuate subducting slab with a SE to E-dipping polarity and the opening of the Alboran back-arc basin. The absence of volcanic activity is a key feature of this stage, although volcanism sporadically reappeared a few million years later. Cordierite-bearing dacite samples in the Mar Menor area have been dated at 18.5 ± 1.6 Ma (Duggen et al., 2004). These anatectic melts were also reported by Zeck et al. (1999) and are explained as melting of pelitic to quartzo-feldspathic clastic rocks in a slab detachment setting. This hypothesis agrees well with the fifth stage, which was characterised by the end of major extension in the Alboran Basin (18-16 Ma) and the onset of slab tear and E-W extension in the Eastern Betics. The occurrence of some calc-alkaline products in the Alboran Sea (ODP 977 at ~ 17.6 Ma; Duggen et al., 2004) was related to active slab retreat in the West Alboran Basin, as proposed by the model.

Most of the volcanic activity of the western Mediterranean region occurred since ~ 15 -14 Ma [hereinafter, all age datings refer to the data of Duggen et al. (2004) and references therein]. During the Middle to Early Miocene, subduction-related magmas with tholeiitic to HK calc-alkaline affinity erupted in different localities: in Ras Tarf (northern Morocco) and Cabo de Gata (southern Spain) at ~ 15 Ma, in the Alboran Sea (ODP 978) around 13 Ma, in Oranie (Northern Algeria) and the Yusuf Ridge (Alboran Sea) at ~ 11 Ma, and in the Alboran Ridge at ~ 10 Ma. This volcanism continued between ~ 9 and ~ 7 Ma, producing volcanic rocks with a typical subduction signature in the Alboran Sea (tholeiitic/calc-alkaline volcanism at ~ 9 Ma) and Northern Algeria (with calc-alkaline affinity, ~ 9 Ma). In southern Spain, calc-alkaline magmas erupted at ~ 8 Ma in Murcia and Mazarron (with sodic alkaline magmas in Picassent; Ancochea and Huertas, 2002). Following stage 6 in Vergés and Fernàndez (2012), this orogenic volcanism is interpreted as a result of partial melting of the mantle wedge related to the

W-directed slab roll-back affecting mainly the western part of the Betic-Rif-Alboran system.

The last volcanic phase occurred in the late Miocene-Quaternary and comprised a wide range of volcanic products. The peculiarity of this period was the eruption of sodic alkaline (anorogenic) magmas soon after emplacement of volcanics with typical subduction-related signature. In northern Morocco, calc-alkaline products erupted at ~7 Ma were followed after only ~1 My by sodic alkaline magmas. Anorogenic magmas erupted from ~3 Ma in northern Algeria (Coulon et al., 2002). Along the Iberian margin, calc-alkaline and HK calc-alkaline magmas were erupted during the late Miocene-Quaternary, especially along the Murcia-Almería coast. Lamproite-like ultrapotassic magmas (8.08-6.37 Ma) were erupted in Vera, Cancarix, Fortuna and Jumilla, some of which were emplaced along transform faults (e.g., the Socovos fault, Pérez-Valera et al., 2013). These rocks are usually interpreted as having originated by partial melting of a strongly metasomatised mantle. Sediments played an important role in the generation of lamproite magmas. If the subducting slab was carrying a large quantity of sediments into the mantle, they would strongly metasomatise the mantle wedge by adding incompatible elements (Ba, Rb, Sr, Th) and K_2O , precipitating clinopyroxene, phlogopite and amphibole either as small veins (Foley, 1992) or disseminated in the peridotitic matrix (Coltorti et al., 2004). On the other hand, Bianchini et al. (2015) linked the source of Spanish lamproites to widespread igneous veining (norite veins) observed in composite mantle xenoliths from Tallante, coupled with very refractory peridotite compositions, known from xenolith occurrences in the Iberian margin (Bianchini et al., 2007). According to those authors, the metasomatism has to account for processes related to Cenozoic and Paleozoic times. The final magmatic input in

southern Spain is represented by the alkaline products of Tallante, erupted between 2.93 Ma and 2.29 Ma (Cebriá et al., 2009).

The Late Miocene-Quaternary volcanic phase in the western Mediterranean area is poorly integrated in the double polarity subduction model. Eruption of calc-alkaline and HK calc-alkaline magmas is in agreement with a mature subduction process (no retreating or active subduction), as proposed by Vergés and Fernàndez (2012) in stage 7. In northern Africa (Morocco and west Algeria), the sodic alkaline volcanic episodes following the typical subduction-related products can be mainly ascribed to three mechanisms: i) sublithospheric flow, as evidenced from mantle anisotropy analysis (Diaz et al., 2010, 2015); ii) slab break-off, as also proposed for the alkaline products of the intra-Apennines volcanism by Bianchini et al. (2008); or iii) splash-plume effect (Davies and Bunge, 2006) due to convective instabilities, as has been proposed by Beccaluva et al. (2011) for Sardinia and Calatrava (southern Spain) volcanism. Furthermore, the occurrence of anorogenic rocks in Tallante is additional evidence of a lateral tear responsible of uplift of the eastern part of the Betics (e.g., Spakman and Wortel, 2004; García-Castellanos and Villaseñor, 2011; Mancilla et al., 2015), as included in the model by Vergés and Fernàndez (2012).

7. Conclusions

We applied a multi-factor statistical approach to a large and well-studied geochemical dataset of volcanic rocks in the central and western Mediterranean area with the aim to reproduce the main petrological characteristics that were previously observed with bivariate diagrams. The main advantage of this method is that it allows investigation of the processes that are responsible for the geochemical signature of the studied volcanic

rock suites in a fast and simple way. This is possible through statistical determination of the main controlling factors constructed by using the original geochemical elements. The method has the drawback of losing part of the information when extracting the factors, thus limiting a detailed interpretation. However, any methodology has its own advantages and drawbacks, hence factor analysis must be considered as a complementary method to classical bivariate diagrams in order to extract all the information contained in geochemical datasets.

Keeping these considerations in mind, the main concluding remarks from the presented study are:

1. After filtering for common geochemical data, the original datasets that encompassed a total of ~7,000 orogenic and ~7,800 anorogenic analyses, were reduced to 283 orogenic and 310 anorogenic volcanic samples.
2. Factor analysis performed using the PCA method reduced the original 36 geochemical parameters, expressed as oxides, elements or isotopic ratios, to seven factors, accounting for ~84% of the variance.
3. Despite the overlap between anorogenic and orogenic geochemical fields, factor analysis discriminates between these two rock types rather well.
4. Lamproites always plot in a separate field, emphasising the great compositional difference among western and central Mediterranean volcanic rocks, and the huge sediment recycling in the mantle responsible for the origin of these exotic rocks.
5. Factor analysis shows mantle metasomatic processes that are responsible for compositional heterogeneities. The controlling factors are related to mantle wedge percolation of fluids from the subducting slab, and extreme sediment recycling.

6. The double polarity subduction model that has been proposed to explain the evolution of the westernmost Mediterranean area is compatible with the timing and type of volcanic rocks and supported by the controlling processes highlighted by the factor analysis.

7. The close relationships between the tectonic processes responsible for the generation of the erupted magmas in the westernmost Mediterranean region and their different geochemical signatures are yet to become well established, thus demanding further multidisciplinary efforts.

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Figure captions

Fig. 1. Sketch map of the Cenozoic orogenic (subduction related) and anorogenic (intraplate) volcanism in Europe, superimposed on the tectonic map of the region modified after Vergés and Sàbat (1999). From west to east: BB, Beni Bousera; R, Ronda; AS, Alboran Sea; MA, Murcia–Almeria; WA, Western Alps; S, Sisco; V, Veneto; SC, Sicily Channel; RP, Roman Province; C, Campania; AA, Aeolian Arc; E, Etna; IL, Insubric Line.

Fig. 2. Timing of anorogenic and orogenic volcanism from the Atlantic Ocean Islands, the Canaries and Madeira, and the Western and Central Mediterranean regions used in this paper for statistical purposes. For references refer to Fig. 4.

Fig. 3. Representative binary plots of the rotated component matrix in terms of F2 vs. F1 (**A**), F3 vs. F1 (**B**) and F5 vs. F3 (**C**) diagrams. See text for a comprehensive explanation.

Fig. 4. Orogenic and anorogenic volcanic rocks of Central (**A**, **C** and **E**) and Western (**B**, **D** and **F**) Mediterranean after the statistical analysis. See text for a comprehensive explanation.

Fig. 5: Representative binary diagrams of the factor scores of Central (**A**, **C** and **E**) and Western (**B**, **D** and **F**) Mediterranean volcanic rocks after the statistical analysis. Samples have been distinguished only on the basis of their age [using different colors following the Commission for the Geological Map of the World (<http://www.ccgmm.org>)] and on the type of geodynamic environment they are linked to. F2 vs. F1 (**A**, **B**), F3 vs. F1 (**C**, **D**) and F5 vs. F3 (**E**, **F**) diagrams. Light blue and light red represent the anorogenic and lamproitic fields, respectively. “Slab dehydration”

indicates that the origin of the volcanic rocks is related to those processes responsible of the dehydration of the subducting slab. When the features of the volcanic rocks need the involvement of sediments in the metasomatic processes of the mantle wedge we used the wording “increasing of sediment recycling”. The dashed arrows indicate the temporal evolution of the Tuscany trend (**A**) and of the samples from Spain (**B**). Anorogenic volcanic data are from: Canary Islands, Madeira, Portugal, Spain, Maghrebian Africa (Morocco), France, Italy (Sardinia, Ustica, Sicily Channel, Etna-Hyblean and Veneto Province) and Germany (Lustrino and Wilson, 2007 and references therein). Orogenic volcanic data are from: Western Alps, France (Corsica – Sisco), Tuscany, Roman Province, Campania, Aeolian Islands, Betics, Alboran Sea, Morocco (Gourougou and Guilliz), Sardinia (Lustrino et al., 2011 and references therein).

Fig. 6. Schematic tectonic maps showing the evolution of the Western Mediterranean since 230 Ma (modified after Jiménez-Munt et al., 2010) Rheic oceanic suture, front of the Variscan belt and major tectonic discontinuities are from Simancas et al. (2005). Modifications of the mantle beneath the Western Mediterranean are probably linked to episodes related to the collision which produced the Variscan Orogeny in the Middle Triassic coupled to the depletion events due to partial melting triggered by subduction of rifting phases (see text for details).

Fig. 7. Topographic map of the Central and Western Mediterranean with the distribution of HP metamorphic complexes along the Betic–Rif system (Internal Betics and Internal Rif), and along the Tellian fold belt in northern Africa (Great and Lesser Kabylies) (modified after Vergés and Fernàndez, 2012). Light orange and orange arrows separated by black dashed line indicate the NW and SE dipping subduction respectively (the double polarity subduction model proposed by Vergés and Fernàndez, 2012). Dashed

white lines indicate other active or inactive faults separating the Alps from the Apennines and the Kabylies. GB stands for Guadalquivir Bank.

ACCEPTED MANUSCRIPT

Table captions

Table 1: Initial Eigenvalues, extraction and rotation of the sums of the squared loadings from factor analysis.

Table 2: Rotated component matrix of the calculated seven factors. Bold represents the best correlation between geochemical elements and factors.

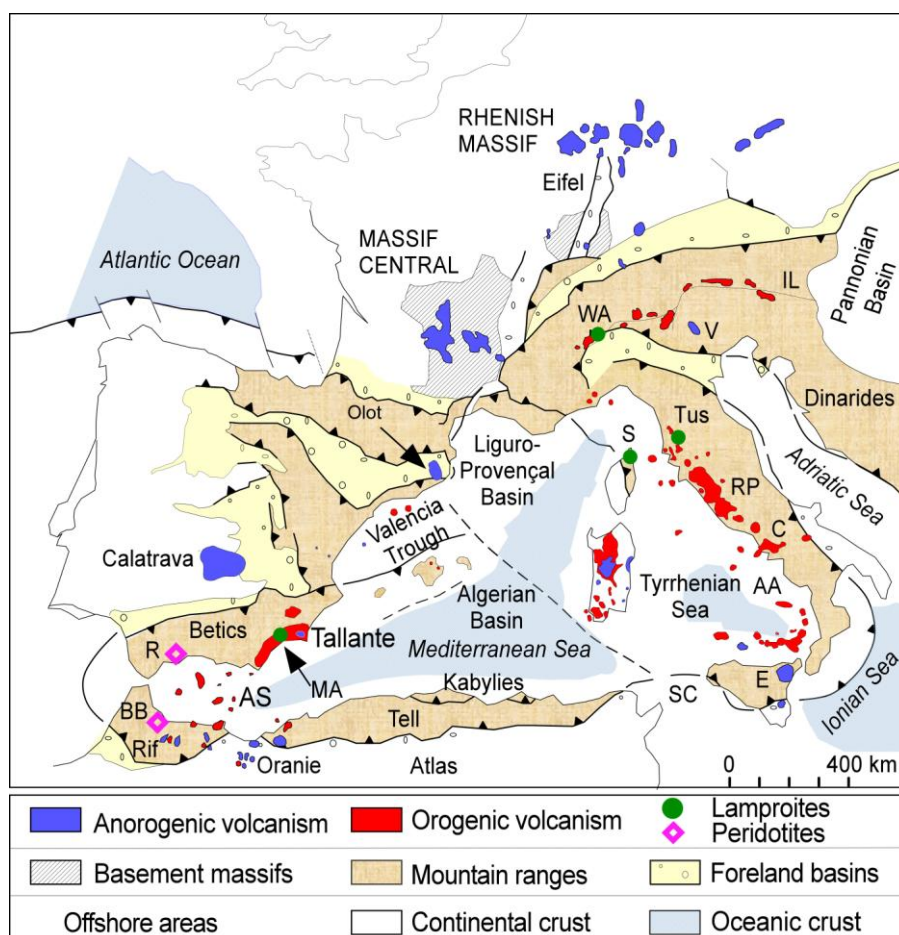


Figure 1

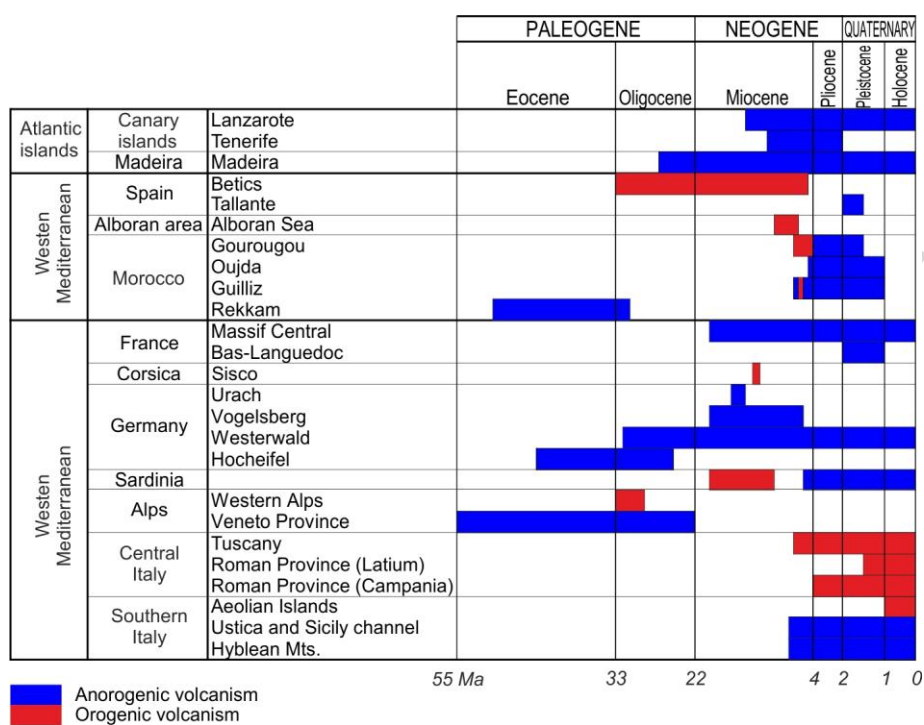
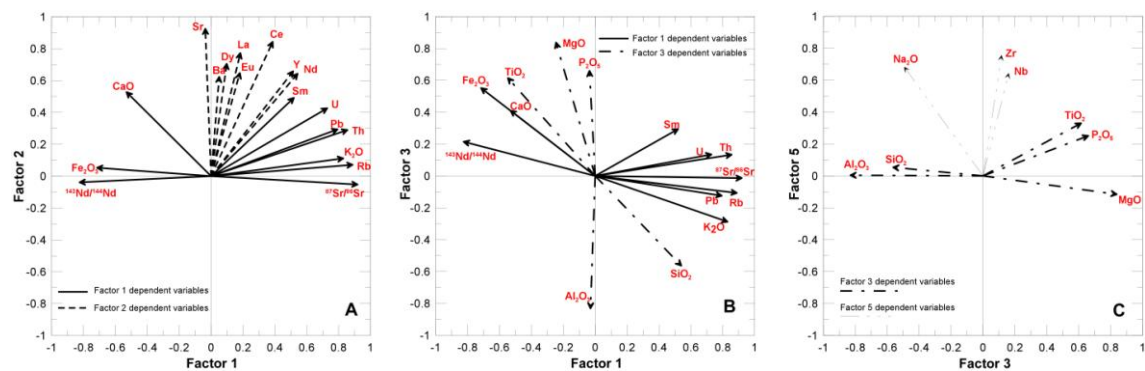


Figure 2

Figure 3



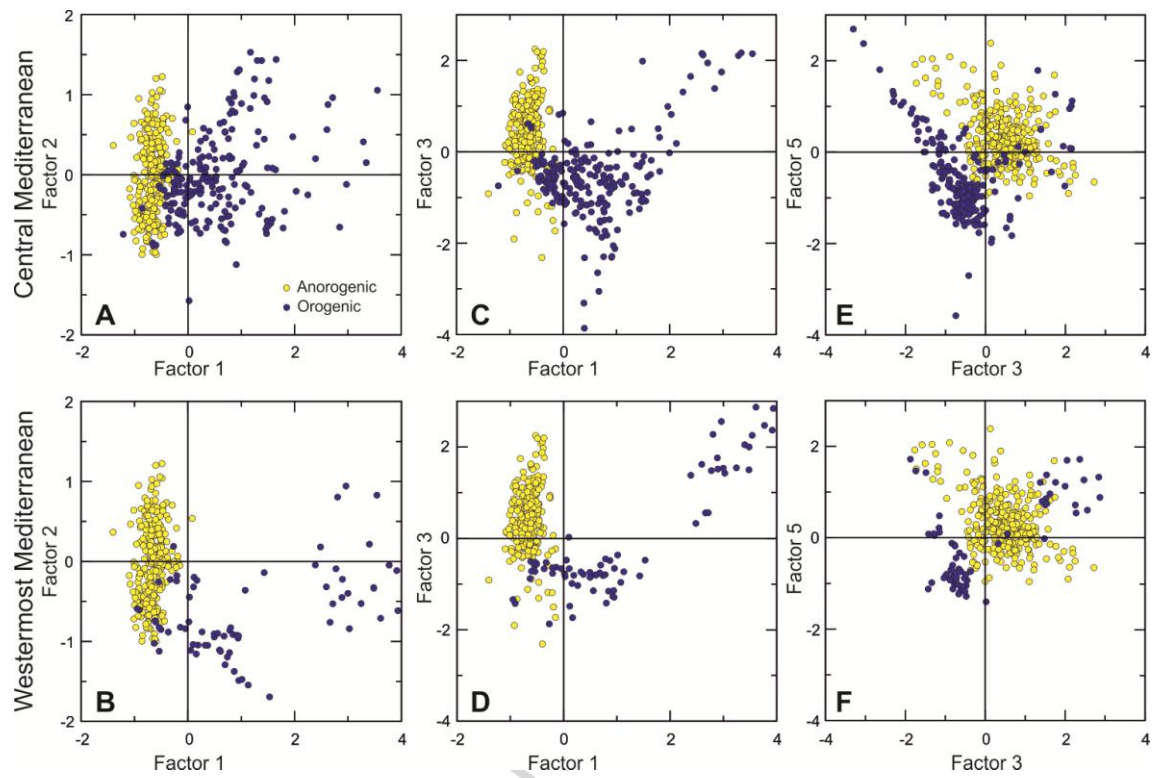


Figure 4

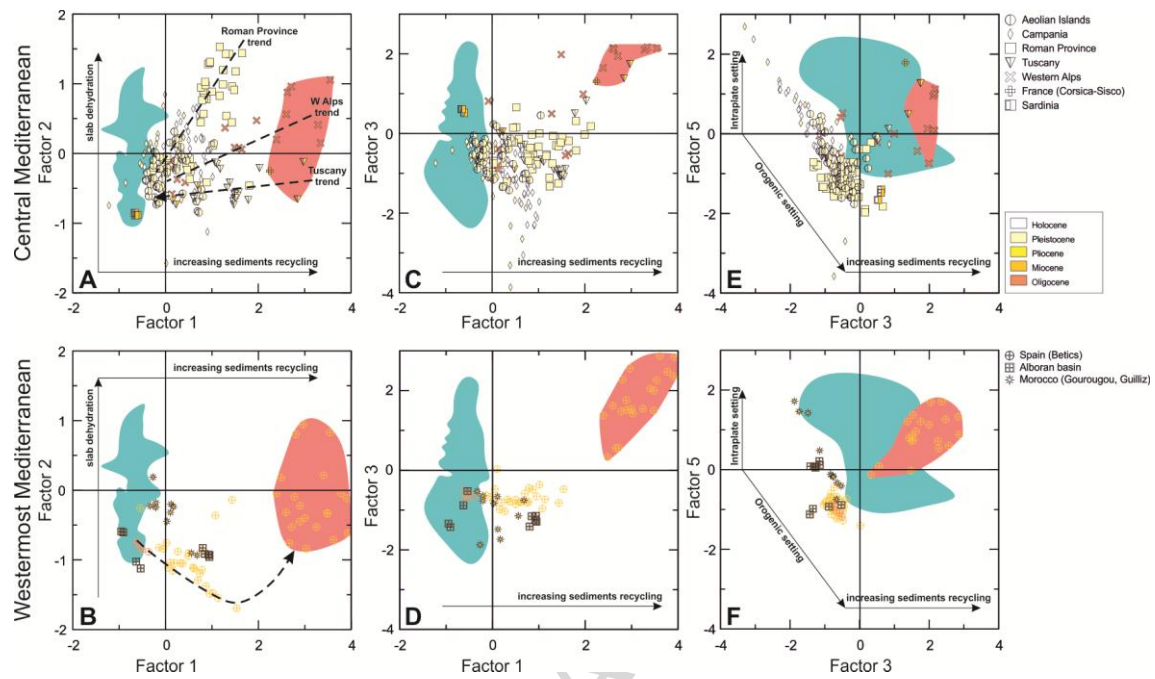
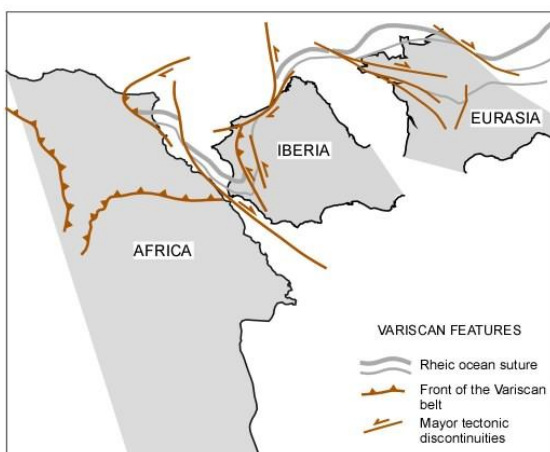
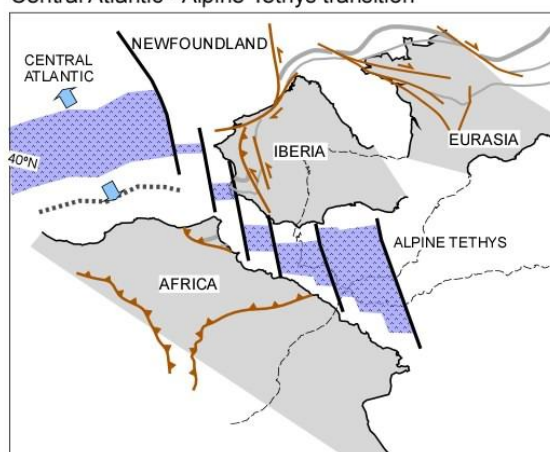


Figure 5

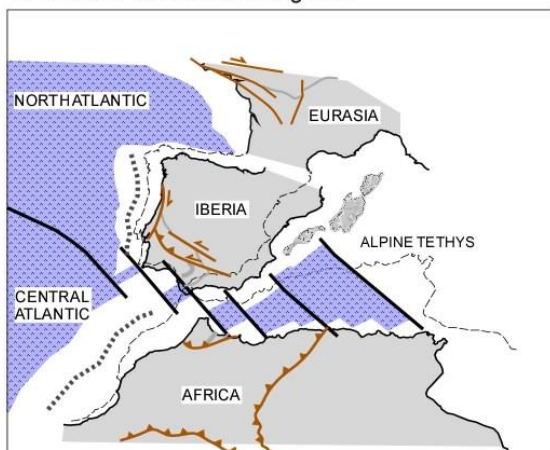
a) 230 Ma Middle Triassic



b) 154 Ma (Chron M25) Late Jurassic
Central Atlantic - Alpine Tethys transition



c) 72 Ma (Chron 32) Latest Cretaceous
Initial Africa northern convergence



d) 8 Ma (Chron 4) Late Miocene
Consumption of Atlantic - Alpine Tethys transition

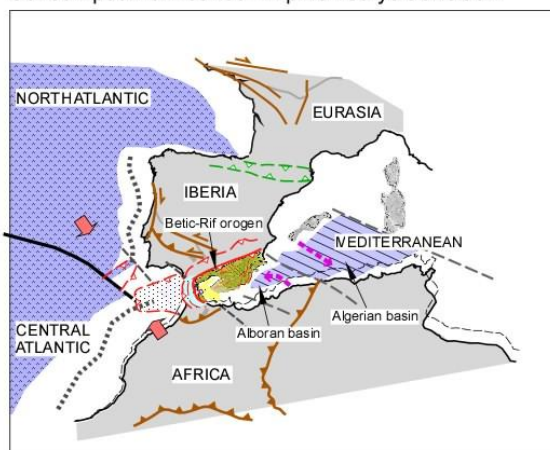


Figure 6

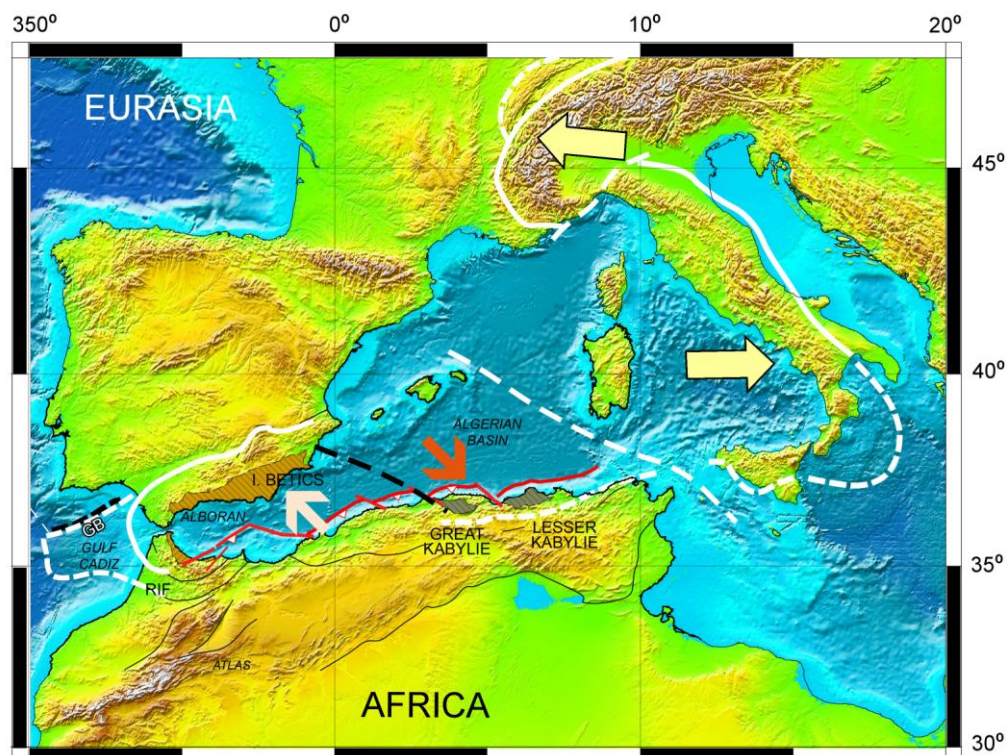


Figure 7

Table 1: Initial Eigenvalues, extraction and rotation of the sums of the squared loadings from factor analysis

Component	Initial Eigenvalues			Extraction Sums of Squared Loadings			Rotation Sums of Squared Loadings		
	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %
1	11.318	31.438	31.438	11.318	31.438	31.438	8.183	22.732	22.732
2	7.595	21.098	52.536	7.595	21.098	52.536	6.697	18.602	41.333
3	3.990	11.084	63.620	3.990	11.084	63.620	4.410	12.249	53.582
4	2.447	6.796	70.416	2.447	6.796	70.416	4.234	11.762	65.344
5	2.125	5.902	76.318	2.125	5.902	76.318	2.302	6.394	71.738
6	1.565	4.347	80.665	1.565	4.347	80.665	2.270	6.306	78.044
7	1.265	3.513	84.178	1.265	3.513	84.178	2.208	6.134	84.178

Table 2: Rotated component matrix of the calculated seven factors. Bold represents the best correlation between geochemical elements and factors.

	Factors						
	1	2	3	4	5	6	7
SiO ₂	.540	-.456	-.563	.023	.054	-.209	-.169
TiO ₂	-.547	.037	.614	.068	.330	.132	.190
Al ₂ O ₃	-.030	-.172	-.831	.018	.004	.182	-.004
Fe ₂ O ₃	-.715	.053	.552	.056	-.033	.166	.142
MgO	-.246	.022	.837	-.115	-.115	.068	.050
CaO	-.531	.527	.410	-.013	-.406	.119	.077
Na ₂ O	-.346	-.019	-.494	-.031	.683	-.086	-.005
K ₂ O	.829	.111	-.287	.084	.134	.026	.096
P ₂ O ₅	-.036	.510	.659	.113	.251	.083	.108
⁸⁷ Sr/ ⁸⁶ Sr	.919	-.053	-.014	.038	-.149	.095	-.123
¹⁴³ Nd/ ¹⁴⁴ Nd	-.825	-.039	.215	-.017	.285	-.074	.204
²⁰⁶ Pb/ ²⁰⁴ Pb	-.361	.082	.180	-.031	.138	-.162	.828
²⁰⁸ Pb/ ²⁰⁴ Pb	.020	.121	.037	-.004	.014	-.122	.915

$^{208}\text{Pb}/^{206}\text{Pb}$	.003	.042	.041	-.073	.000	.974	-.100
$^{207}\text{Pb}/^{206}\text{Pb}$	.016	.034	.030	-.070	-.006	.969	-.156
Rb	.888	.071	-.107	.128	.055	.028	.072
Sr	-.036	.925	.059	.065	-.136	.021	-.018
Ba	.515	.658	.170	.029	-.002	.055	-.136
Y	.051	.623	-.236	.406	.099	-.085	.296
Zr	.449	.231	.116	.041	.757	.081	.140
Nb	-.317	.526	.161	.053	.644	-.027	.135
La	.184	.772	.038	.408	.173	-.012	-.015
Ce	.387	.843	.083	.173	.158	-.002	.096
Pr	.394	.529	.204	.625	.016	.017	-.043
Nd	.543	.643	.233	.175	.151	.051	.103
Sm	.519	.491	.294	.345	.163	.069	.052
Eu	.180	.647	.443	.371	.232	.195	.105
Tb	.221	.205	.095	.843	-.026	.050	-.042
Dy	.098	.705	.026	.366	.185	.003	.353
Ho	-.059	.218	.028	.885	-.036	-.048	.053
Tm	.098	.091	-.097	.933	-.033	-.115	-.074

Yb	.112	.450	-.498	.334	.209	-.204	.358
Lu	.061	.236	-.402	.666	.246	-.106	.120
Th	.855	.290	.133	.148	.137	-.065	-.045
Pb	.793	.294	-.124	.122	-.029	.047	.018
U	.729	.426	.135	.156	.026	-.168	-.124

Highlights

1. Factor analysis is applied to geochemical database with orogenic and anorogenic rocks
2. Mantle heterogeneities are highlighted in both Central and Western Mediterranean
3. Heterogeneities are mainly due to slab-released fluids and sediment recycling
4. Double subduction polarity model is compatible with the Cenozoic volcanic petrology