Cretaceous ⁴⁰Ar/³⁹Ar detrital mica ages in Tertiary sediments shed a new light on the Eo-Alpine evolution.

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Abstract: Clastic sediments deposited in the syn-orogenic Tertiary Piedmont Basin in northwest Italy represent the depositional counterpart of the cooling/exhumation and erosion of Western Alpine rocks over the last 30-35 My. 40Ar/ 39 Ar analyses of detrital white micas from Oligocene-Miocene sediments and present-day river sands show, in addition to younger Tertiary age groups, a wide range of Cretaceous ages. Pronounced well defined Late Cretaceous (~70-90 Ma) and Early Cretaceous (~105-120 Ma) age clusters are recorded in Lower to Upper Miocene sediments, forming discrete age groups with a contribution to the total detrital population of as high as 58%. This age pattern of discrete age peaks is remarkably constant and can be followed up-sequence through different formations spanning a time interval for sedimentation of >20 Myrs. Our new detrital mica ages may be the result of either excess Ar in the source rock, as commonly assumed for HP mica Ar ages from the internal western Alps, or of inherited Ar, and consequently be representative of real geological events. The first scenario would imply that incorporation of excess Ar in minerals can lead to non-random detrital age populations which could then mistakenly be interpreted as representative of real geological events. The second scenario would imply that during the last Eocene thermal event, pre-existing micas were only partially overprinted and the presence of older ages are the result of real Cretaceous metamorphic events of the Western Alpine orogen. We argue that our new data derived from the sedimentary record, in particular from the time interval from Serravallian to Present, cannot easily be explained as simply being due to incorporation of excess argon. We therefore interpret these ages to be representative of cooling following major metamorphic events in the Alpine orogen. The new argon data from the sediments in combination with the data from the rocks exposed in the orogen today point to a complex Mesozoic history of the internal Alpine orogen.

INTRODUCTION

Although the Alpine chain has been widely studied, many ongoing issues regarding the timing of the poly-metamorphic history, the pattern of exhumation and the possible effects of a Cretaceous HP metamorphic event are still strongly debated (England and Molnar, 1990; Rubatto and Hermann, 2001). For instance, several authors have dismissed the Cretaceous signal and have attributed ages ranging around 80-120 Ma (⁴⁰Ar/³⁹Ar dating on white micas) to incorporation of excess ⁴⁰Ar (e.g. Arnoud & Kelley, 1995; Reddy et al., 1996). For this reason recent paleogeographic reconstructions suggest that subduction, leading to high pressure and ultrahigh pressure metamorphism in the Alpine orogen, occurred no earlier than Late Cretaceous time (e.g. Agard et al., 2002; Reddy et al., 2003) and dismiss the possibility of an older history preceding this. However, regardless of any isotopic constraint, the only upper limit of the oceanic domain and lower limit for the onset of the subduction is given by the presence of Callovian-Oxfordian (~165-154 Ma) Alpine/Apennine supra-ophiolitic sediments (e.g. Wever et al., 1987), which leaves room for interpretations that invoke a more extended Cretaceous metamorphic history for the Western Alps. The purpose of this paper is to investigate the validity of Cretaceous events by looking at the geochronological signal recorded by clastic sediments derived by the erosion of the Western Alpine orogen.

Continuing exhumation and erosion prevent us from assessing the rock record in orogenic belts at any time before the present. This record is available, however, in the form of sediments in syn-orogenic clastic sedimentary basins such as the Tertiary Piedmont Basin (TPB). Thus, constraints on processes of cooling and exhumation of the orogen can be obtained from sediment petrography. In this context detrital mineral geochronology is a unique technique because it provides quantitative information on cooling/exhumation and erosion events in the surrounding orogen as it develops through time.

The TPB (Figure 1), located within the Internal Western Alpine Arc, is one of the main sedimentary basins collecting the products of the cooling/exhumation and subsequent erosion of Western Alpine rocks over the last ca. 30 My (Carrapa, 2003c and references therein). Oligocene sediments deposited in the TPB are derived



Figure 1. Tectonic map of the Alps showing the major nappe complexes (the Austro-Alpine, the Penninic with the Ophiolitic domain and the Helvetic), modified from Bigi *et al.* (1983). AGM: Argentera Massif, DM: Dora Maira, GP: Gran Paradiso. Square: study area.

mainly from the Ligurian Alps to the south (Barbieri *et al.*, 2003), while Miocene sediments are derived from westernnorth-western Alpine rocks including the Internal Western Alpine Massifs and possibly the Sesia Lanzo zone (Cara pa *et al.*, 2003c). 40 Ar/ 39 Ar analysis on detrital white micas has been performed on samples from the entire TPB clastic sequence ranging from Late Eocene – Present, providing a complex picture of age signals caused by cooling/exhumation events. In this paper the discussion will focus on the importance of the Cretaceous ages and we refer to Carrapa (2002) for discussion of other facets of the database.

GEOLOGICAL SETTING AND EVOLUTION OF THE TPB

The Tertiary Piedmont Basin (TPB) is located within the Internal Western Alpine Arc at the junction of the Alpine and the Apennine thrust belts sealing at present their tectonic relationships (Figure 1). From Late Eocene-Early Oligocene until Late Miocene times the TPB accommodated sediments from the erosion of the surrounding source areas (Internal Western Alps). The TPB is bounded to the south by the Ligurian Alps (including the Voltri Group), which form the easternmost segment of the Western Alpine Arc, to the west by the rest of the Western Alpine domain, to the north and north-east by the Po Plain and to the east by the Northern Apennine (Figure 1).

In the study area the clastic sequence contains up to 4 km Oligocene-Upper Miocene sediments (Figure 2a, 2b, 2c). Subsidence in the TPB started in the latest Eocene-Early Oligocene and caused the progressive drowning of the Ligurian Alps. The onset of sediment deposition occurred mostly in the Early Oligocene (Dela Pierre *et al.*, 1995). Sedimentation continued until Late Miocene time with strong subsidence during the Langhian (Carrapa *et al.*, 2003a; Gelati *et al.*, 1993) followed by recent uplift (e.g. Lorenz, 1984) allowing the entire stratigraphic series to crop out at present.

The first sediments deposited in the study area are of alluvial and near-shore environmental settings (i.e. the Molare Formation) (Lorenz, 1979; Lorenz, 1984; Gelati et al., 1993; Gelati and Gnaccolini, 1996; Gelati and Gnaccolini, 1998; Mutti et al., 1999). Petrographical, geochronological and sedimentological analyses of the Molare Fm. indicate a provenance of these sediments from southern areas (Ligurian Alps; e.g. Gnaccolini, 1974; Barbieri et al., 2003 and references therein). At the end of the Early Oligocene, an increase of subsidence (Dela Pierre et al., 1995) occurred coevally with the deposition of a transitional/shallow marine marly/sandy sequence known as the Rocchetta Formation (Upper Oligocene-Aquitanian; Artoni et al., 1999; Gelati and Gnaccolini, 1996; Mutti, 1985 and references therein). The contact between the Molare Fm. and the Rocchetta Fm. is progressively younger from NE to SW (e.g. Fravega et al., 1994; Gelati et al., 1993; Vannucci et al., 1997).

The Early-Middle Miocene was generally characterised by more widespread basinal conditions (e.g. Gelati *et al.*, 1993) and the deposition of hemipelagic sediments. The entire central area was characterised by silty/sandy sequences within which several different depositional bodies can be distinguished on the basis of the silt/ sand ratio (Rocchetta-Monesiglio Group; Cortemilia and Paroldo Fms.; Gelati and Gnaccolini, 1998; Gnaccolini *et al.*, 1990). Petrographic and geochronological data together with paleocurrent directions indicate a contribution from



Figure 2a. Geological framework of the study area modified after Gnaccolini *et al.* (1998). 1) Pliocene to Recent deposits; 2) Messinian deposits; 3) Langhian to Tortonian siliciclastics and carbonates shelf to slope deposits; 4) Late Burdigalian to Tortonian mainly turbidite succession (only Burdigalian in the eastern sector of the figure); 5) Late Oligocene to Burdigalian turbidite systems and hemipelagic mudstones; 6) Late Eocene to Early Oligocene deposits: (a) alluvial to coastal conglomerates, shallow marine sandstones and hemipelagic mudstones, (b) slope and base-of-slope, resedimented conglomerates and (c) mainly turbidite; 7) Late Eocene to Tortonian siliciclastic deposits of the northwestern Apennines-Monferrato-Torino Hill wedge; 8) Alpine and Apennine allochthonous units; 9) depocentre axis of the Plio-Quaternary basins; 10) buried thrust-front of the Torino Hill-Monferrato-northwestern Apennine wedge; 11) buried, southvergent backthrusts of the Monferrato, active from Messinian onward; 12) buried, pre Burdigalian backthrusts of the Western Alps (as inferred from Roure *et al.* 1990; 13) faults: SV Sestri-Voltaggio, VVL Villalvernia-Varzi line. I: Bagnasco-Ceva-Bastia Mondovi' transect; II: Millesimo-Monesiglio-Somano transect; III: Dego-Torre Bormida-Alberetto della Torre transect; IV: Spigno Monferrato-Cessole transect; V: Montechiaro d'Acqui; VI: Cavatore; VII: Visone. The square corresponds to the area of the paleocurrent study of Gnaccolini & Rossi (1994).



Figure 2b. Oligo-Miocene depositional sequences (A, B1-B6, C1-C6) in the study area modified after Gelati *et al.* (1993). 1: mudstones, locally with thin-bedded turbidites (a); 2: turbidite systems (sand/mud ratio from very high to = 1; locally conglomerates); 3: resedimented ophiolite breccia; 4: olistolith-bearing pebbly mudstones; 5: shallow-water carbonates; 6: alluvial conglomerates, coastal sandstones and conglomerates, fresh-water mudstones (a); 7: Pre-Cenozoic rocks; 8: sequence boundary; 9: fault. The area into the square is mainly based on Cazzola *et al.* (1981), Cazzola & Sgavetti (1984) and Cazzola & Fornaciari (1990). The ages of the unconformities/sequence boundaries are based on planktonic foraminifers and calcareous nannofossils and are referred to the correlation with the boundaries (Ma) of the third order global cycles of Haq *et al.* (1988).



Figure 2c. Stratigraphic schematic scheme modified after Gelati (1968).

western Alpine rocks to these sediments (Gnaccolini and Rossi, 1994; Carrapa, 2002).

During the Middle-Late Miocene different formations developed in the TPB. The eastern part of the TPB was characterised by shallow-water shelf sedimentation (Caprara et al., 1985; Ghibaudo et al., 1985) while basinal conditions prevailed elsewhere. This allowed the deposition of tabular turbidites and interbedded hemipelagites constituting the Murazzano Fm., the Cassinasco Fm., the Cessole Marls and the Lequio Fm. (Gelati et al., 1993). Petrographic and geochronological data together with paleocurrent directions indicate a main source area for these sediments located in western north-western areas including the Internal Western Alpine Massifs and possibly the Sesia Lanzo zone (Gnaccolini and Rossi, 1994; Carrapa et al., 2003c). The Miocene succession is unconformably overlain by alluvial and evaporite sediments recording the Messinian Mediterranean salinity crisis (Ghibaudo et al., 1985).

GEOLOGICAL SETTING OF THE AREAS SUR-ROUNDING THE TPB

Western Alps

During the Mesozoic the break-up of the Pangea supercontinent led to the formation of the Tethys Ocean between the European continent to the north (Eurasia) and the African continent to the south (Frisch, 1981; Platt *et al.*, 1989). The portion of Tethys between the north- eastern margin of the African continent (Adria) and the southern margin of Eurasia was the Ligure-Piemontese Ocean. The Alpine chain formed as a consequence of the Mesozoic subduction of the Ligure-Piemontese Ocean and of the subsequent collision between Adria and Eurasia.

The Western Alpine arc consists essentially of three nappe complexes: the western Austroalpine, the Penninic (upper, middle, lower) and the Helvetic (Ultrahelvetic, Helvetic and Dauphinois units) (Figure 1). All nappe complexes (except the upper Penninic) comprise a crystalline basement covered by Mesozoic and Tertiary sediments. The upper Penninic belongs to the oceanic domain and includes the Piedmont ophiolitic suture. The middle (e.g. Briançonnais domain) and lower Penninic together with the Helvetic belong to the European margin while the Austroalpine domain belongs to the Adria margin, which was separated from Eurasia during the Jurassic by the Ligure-Piemontese Ocean.

In general the Western Alps experienced a complicated poly-metamorphic evolution reflecting continued convergence, hence the importance of establishing the complete sequence of metamorphic events in time. In the following the most important HP-UHP metamorphic phases are summarised (see also Agard *et al.*, 2002 for a review). In sequence, from the top of the nappe stack to the bottom:

-Western Austroalpine: Eo-Alpine (~130?-110?/65 Ma) (Polino *et al.*, 1990; Gebauer, 1999 for a review) eclogitic metamorphism (e.g. Sesia Lanzo and Dent Blanche; Dal Piaz *et al.*, 1983; Inger *et al.*, 1996; Cortiana, *et al.*, 1998; Rubatto *et al.*, 1999; Ruffet *et al.*, 1995) and Meso-Alpine overprint (Late Eocene-Early Oligocene) greenschist metamorphism (Reddy *et al.*, 1996; Dal Piaz, 1999);

-Piedmont ophiolitic suture: Eo-Alpine (~110?-70 Ma) (Polino *et al.*, 1990; Gebauer, 1999; Schwartz *et al.*, 2000) eclogitic and blueschist metamorphism (e.g. Schistes Lustrés; see also Agard *et al.*, 2002; Takeshita *et al.*, 1994) and Meso-Alpine (Late Eocene-Early Oligocene) greenschist and UHP (e.g. coesite-glaucophane assemblages) overprint (e.g. Zermat-Saas zone; Dal Piaz, 1999; Dal Piaz *et al.*, 2001; Reinecke, 1998; Rubatto *et al.*, 1998; Dûchene *et al.*, 1997; and references therein).

Upper Penninic domain: Eo-Alpine (~110?-60 Ma) (Polino *et al.*, 1990; Gebauer, 1999 and references therein) eclogitic metamorphism and greenschist Meso-Alpine (~35 Ma) overprint (e.g Dal Piaz 2001). Coesite-pyrope assemblages have been recognised in the Dora Maira Massif and interpreted as the result of Meso-Alpine UHP metamorphism followed by fast cooling/exhumation during the Eocene (e.g. Avigad, 1992; Chopin, 1984; Cortiana *et al.*, 1998; Gebauer *et al.*, 1997; Hurford *et al.*, 1991; Monié and Chopin, 1991; Rubatto *et al.*, 1999; Rubatto and Hermann, 2001).

-Lower Penninic domain: Eo-Alpine–Meso-Alpine eclogitic to blueschist metamorphism (Bocquet *et al.*, 1984), and Meso-Alpine greenschist to amphibolite facies metamorphic overprint (Dal Piaz, 1999).

The meaning of Eo-Alpine ages detected in the Western Alps, especially in the Internal Western Alpine Massifs

(e.g. Dora Maira; Monte Rosa; Chopin, 1984; Monié and Chopin, 1991; Paquette et al., 1989; Scaillet et al., 1992; Scaillet et al., 1990) is still a matter of debate. Many authors have dismissed the geological meaning of these ages by attributing them to the presence of excess 40 Ar (e.g. Arnaud and Kelley, 1995; Kelley et al., 1994; Ruffet et al., 1995). However, thermochronometers such as U/Pb on zircons, Rb/Sr on whole rock and zircon fission tracks (Oberhänsli et al., 1985; Paquette et al., 1989; Cortiana et al., 1998; Gebauer et al., 1999; Vance, 1999; Schwartz et al., 2000) have recorded complementary data suggesting that real geological events occurred in the Cretaceous in the Western Alps. In particular, Paquette et al. (1989) proposed an Eo-Alpine eclogite-facies metamorphism phase dated between 95 and 120 Ma in the Monte Rosa, Dora Maira Massifs and Sesia Lanzo zone. Their results, obtained by U-Pb zircon, Rb-Sr on whole rock, apatite and phengite and whole rock Sm-Nd on meta-acid rocks, are concordant with ${}^{40}\text{Ar}/{}^{39}\text{Ar}$ plateau ages on phengite (105-110 Ma) obtained by Monié (1984) and Chopin & Monié (1984) on the same rock samples. Also Cortiana et al. (1998) suggested a Late Cretaceous HP event for the Austroalpine-Piedmont domain. Their interpretation is based on concordant Rb/Sr and ⁴⁰Ar/³⁹Ar plateau ages on phengitic micas. These data already suggest that a possible link between Cretaceous ages to HP metamorphism in the Alps is not unrealistic.

Ligurian Alps

The Ligurian Alps form the south-westernmost segment of the Alpine collisional belt (Figure 3) and comprise different domains (Provençal, Dauphinois, Briançonnais and Piemontese domains) belonging to the paleo-European continent and the oceanic crust of the Ligure-Piemontese domain (Figure 3) (Vanossi *et al.*, 1984; Vanossi, 1991).

The Provençal-Dauphinois domain consists of a crystalline basement and a Permo-Cenozoic cover. The pre-Upper Carboniferous basement (gneisses, amphibolites, granitoids) and cover units crop out at present in the Argentera-Mercantour Massif.

The Briançonnais domain is a tectonic complex, which records the pre-Alpine history of the European crust, related mostly to the Variscan orogenic cycle (Devonian- Carboniferous) and Late Paleozoic-Mesozoic sedimentary evolution (Vanossi *et al.*, 1984; Cortesogno *et al.*, 1993). The Briançonnais domain and the Provençal Dauphinois domain both comprise a crystalline basement (i.e. Variscan Crystalline Massifs) and Permo-Cenozoic covers.

The Piemontese domain consists of either only Meso- Cenozoic covers or crystalline rocks (e.g. Valosio Crystalline Massif).

The Ligure-Piemontese domain consists of ophiolitic units (i.e. Montenotte Nappe, Voltri Group, Erro-Tobbio, Sestri Voltaggio) and flyschoid sequences. The Montenotte



Figure 3. Tectonic sketch of the Ligurian Alps with reconstructed paleogeographic position of the different domains modified after Vanossi *et al.* (1984).

Nappe is derived from the transitional domain between the ocean and the palaeo-European continental margin and experienced subduction metamorphism mainly under blueschist conditions during Alpine orogenesis (Messiga, 1984). The Voltri Group is a meta-ophiolite massif, which represents a remnant of Piedmont-Ligurian oceanic crust with its deep-sea cover. The Piedmont-Ligurian Ocean formed as a result of spreading in the Jurassic between Europe and Africa, and closed during Cretaceous-Paleogene Alpine convergence between the two continental blocks. The Voltri Group records a complex retrograde metamorphism from eclogitic (Alpine subduction metamorphism) to greenschist conditions (Messiga, 1984).

Few geochronological data available on the Ligurian Alps concern mainly the Briançonnais domain and the Voltri Group (Del Moro *et al.*, 1981; Hunziker *et al.*, 1992). The Briançonnais domain is characterised by Variscan ages (Del Moro *et al.*, 1981; Hunziker *et al.*, 1992), while the Voltri Group exhibits mainly Eocene- Oligocene ages (Barbieri *et al.*, 2003). Jurassic and Cretaceous ages around 180-120 Ma have been recorded by zircon fission track (ZFT) analysis in the Briançonnais-Piemontese domain and in the Helmintoid Flysch of the Ligurian Alps (Vance, 1999).

METHOD

⁴⁰Ar/³⁹Ar geochronology has been applied to detrital phengites from over 60 samples from selected stratigraphic units of the TPB, ranging from Oligocene to Tortonian in age, and also present-day river sands. The general chemistry of the studied samples is shown in Figure 4 and we refer to Barbieri et al. (2003) for an extended discussion on the chemistry of the Molare Formation. An average of 7 samples were considered for each formation and 2 samples for each river. Single fusion analysis has been applied on ca.10 single crystals (ranging between 250-500 mm size) from each selected sample, for a total of over 500 individual experiments (Figure 5a). Step heating has been applied to 10 single grains ranging between 500-1000 mm in size, derived from different formations and rivers, to check the internal distribution of radiogenic ⁴⁰Ar in each sample. Only experiments concordant within 95% confidence intervals, i.e. MSWD <2.5 have been used to derive plateau ages. The ages obtained on TPB clastic phengites are interpreted to represent the time of isotopic closure during cooling of the crystalline source through 350-420∞C.

The ⁴⁰Ar/39Ar experiments were carried out with the VULKAAN laserprobe facility at the Isotope Geology Laboratory of the Vrije Universiteit in Amsterdam following laser extraction and mass spectrometry methods for this facility described by Wijbrans *et al.* (1995). Samples and standards, individually wrapped in Al- and Cu- foils were stacked into quartz tubes. The quartz tubes were irradiated for 7 hrs. in the cadmium–lined CLICIT facility of the TRIGA reactor of the Oregon State University Reactor Center. Correction factors for interferences of Ca and K isotopes were 0.000673 for 39Ar/37Ar, 0.000264 for 36Ar/37Ar and 0.00086 for ⁴⁰Ar/39Ar respectively. These



Figure 4. Chemical data of selected samples from the TPB clastic sequence. N=number of analyses (10 for each sample). For a detailed discussion on chemistry of the Molare Formation and for analytical procedures we refer to Barbieri *et al.* (2003).

values were determined using zero age K-feldspar and anorthite glass. After irradiation, a J curve was derived for individual samples by interpolation between 5 single fusion experiments on every flux monitor. As flux monitor standards for this project we used TCs (Taylor Creek sanidine) and DRA sanidine (Steenbrink et al., 1999), with an age of 28.34 ± 0.16 Ma and 25.26 ± 0.14 Ma respectively. These values are compatible with the set of (Renne et al., 1998), based on biotite GA1550 (at K/Ar age of 98.79 \pm 0.69 Ma). In the present study, system blanks were determined after every 5 unknowns. The unknowns were corrected for the interpolated blank at the time of analysis of the unknown and the 2s error on the blank was carried over for the error calculation of the unknown. 40 Ar intensities for the analysed samples were in the order of >100 times the blanks (see Wijbrans 1995 for further details on mass spectrometer sensitivity). For the whole dataset we refer to (Carrapa, 2002).

Probability distribution diagrams (Sircombe, 1999; Sircombe, 2000) have been used to identify the main populations of detrital ages present in different formations of the TPB clastic infill and present-day river sands. The probability distribution curves provide the most frequent ages characteristic of a given sample taking into account the analytical uncertainty of each



Figure 5a. Plot of detrital-mineral 40 Ar/39Ar ages vs. minimum stratigraphic ages characteristic of each formation and present-day river sands after Carrapa *et al.* (2003b).

analysis (e.g Sircombe, 2000). For some formations Rocchetta- Cassinelle, Cortemilia-Paroldo, (e.g. Murazzano-Cassinasco) results have been combined, as these formations contain similar sedimentological patterns (similar depositional environment and/or similar petrographic compositions and paleocurrent directions) and are synchronous. The approach of using most frequently occurring ages in complex populations to discriminate real geological signals, has also been successfully applied in cases where complex apparent age spectra are produced by mixing of different components in a single sample (e.g. Forster and Lister, 2003; Najman et al., 2001; Sambridge and Compston, 1994). Complex patterns of ages in the past often have been discounted as un-interpretable. However, in many geological environments mixtures of signals are produced that can be understood following detailed analysis of the dataset. Single grain dating techniques represent a key advancement in tackling such complex systems. Additional information can be obtained in favourable cases by incremental heating or multi-spot fusion analysis of carefully characterised samples (Forster and Lister, 2003).

RESULTS

Our data clearly show the presence of several distinct Cretaceous age populations, which can be followed through different formations up-sequence for over 30 My as age clusters or age peaks in cumulative probability diagrams (Figure 5b). The proportion of the sample set that produced Cretaceous provenance ages in the sediments increases from 13% in the Oligocene up to 58% in Late Miocene times. The 68-75 Ma, the 82-92 Ma and the 107-120 Ma populations are the most persistent. Ages between 93 and 106 Ma appear as transient signals in the detrital mineral age population (Figure 6). Within the persistent populations two very distinctive peaks can be traced, one at 70 Ma and the other at 120 Ma. These ages are also supported by step heating analyses, which gave plateau ages in the Rocchetta (D61b) and Cassinasco fms. (D40, D50) of 108.7 ± 1 Ma and 105.7 ± 2 Ma, 77.6 ± 1 Ma respectively (Figure 6). Also, the 107-120 Ma age population is remarkably close to ⁴⁰Ar/39Ar plateau ages on white mica and Rb/Sr ages on whole rocks measured in the Internal Western Alpine Massifs and in the Sesia Lanzo zone (i.e. of rocks exposed at the surface today) (Hunziker, 1970; Oberhänsli *et al.*, 1985; Paquette *et al.*, 1989). Ages in the range of 70- 90 Ma have also been widely recorded by different thermochronometers (e.g. U/Pb on zircons and 40 Ar/39Ar and Rb/Sr on white mica), all over the Western Alps (e.g. Chopin, 1984; Inger *et al.*, 1996; Monié and Chopin, 1991; Oberhänsli *et al.*, 1985; Ruffet *et al.*, 1997).

Excess 40 Ar or inherited 40 Ar?

One of the fundamental assumptions underlying the K-Ar and 40 Ar/39Ar dating methods is that the system does not contain 40 Ar that the time that the clock starts, i.e. at the time of crystallization of a mineral or the time of cooling through its closure temperature, and thus the calculated age can be related to the event that we intended to study. If the system contains extraneous argon at this time, then ages will be measured that are older than the true age of crystallization or cooling (Dalrymple and Lanphere, 1969). The extraneous, i.e. the non-radiogenic 40 Ar component may have various sources. For the present discussion two components are relevant: excess 40 Ar and inherited 40 Ar.

Following Dalrymple and Lanphere (1969) and Kelley (2002) excess ⁴⁰Ar is represented by parent-less radiogenic argon incorporated into a mineral during crystallisation, or introduced into the mineral lattice by subsequent diffusion or occluded within fluid or melt inclusions within the mineral. Thus, excess ⁴⁰Ar is controlled by diffusion and fluid advection processes transporting 40Ar during metamorphic events from areas where minerals break down to areas where new minerals crystallize. Transport distances of excess, i.e. parentless, ⁴⁰År may range from micrometres (Harrison and McDougall, 1981) to hundreds of metres (York et al., 1981). By definition, in the case of excess 40 Ar the relation between radiogenic parent isotope 40K and radiogenic daughter isotope 40 Ar is broken and thus no interpretable age information can be obtained. Excess argon may be deeply or superficially bound in the crystal lattice of minerals but is also likely to reside in fluid inclusions and solid inclusion within minerals.

Inherited argon is defined as the ⁴⁰Ar component, essentially radiogenic, present in a rock or mineral sample



Figure 5b. Relative Probability distribution diagrams of Cretaceous ages discussed in this study (see text for explanation). For the whole dataset see Figure 5a and refer to Carrapa (2002).

by physical contamination from older material. This definition implies that in the case of inherited argon, the relation between radioactive parent 40K and radiogenic daughter, 40 Ar, is maintained (e.g. Dalrymple and Lanphere, 1969; Wijbrans and McDougall, 1986; Singer *et al.*, 1998). In the case of inherited argon in metamorphic rocks, the apparent age is produced, in part, by the decay of 40K, before the last cooling event. Thus, anomalously old ages caused by a component of inherited argon will never exceed the protolith age of the rock.

While excess Ar leads to age spectra, which cannot be interpreted in terms of the effects of real geological events, inherited Ar will produce age spectra that are influenced by mixing components of different ages, which however, under favourable circumstances can be interpreted in terms of the compounded effects of geological events (e.g. Wijbrans and McDougall, 1986; Villa, 1998; Forster and Lister, 2003).

DISCUSSION

The presence of excess ⁴⁰Ar has been recognised especially in high-pressure metamorphic rocks (e.g. Arnoud and Kelley, 1995; Inger, 1996; Scaillet, 1996; Ruffet, 1997; Sherlock and Kelley, 2002). Under the assumption of mobility of argon in a crystal lattice by volume diffusion, excess ⁴⁰Ar is expected to produce age spectra that are

defined at the lower limit by a potentially real geological age while the upper limit is unconstrained and tends to go to values without any geological meaning (e.g. Pankhurst *et al.*, 1973; Harrison and McDougall, 1981). Diffusion gradients with high apparent ages at the grain boundaries, decreasing toward the crystal cores are typical for excess argon (Reddy *et al.*, 1996; Sherlock and Kelley, 2002). Grains containing inherited argon on the other hand, are more likely to show the reverse pattern with higher ages found in the cores of the grains.

Different scenarios for incorporating excess Ar, briefly described as follows, have been proposed (e.g. Arnoud and Kelley, 1995).

-Incorporation of a large quantity of excess argon during HP peak metamorphism, followed by a partial redistribution of argon during retrogression. This process would produce a range of ages in different units as a result of the non-uniform later partial redistribution of Ar during retrogression.

-Incorporation of argon during the HP peak, but also during the retrogression path. This process, as well, would create a wide range of ages due to the different retrogression paths experienced by different Alpine units.

Examples of excessively high ⁴⁰Ar/³⁹Ar spot ages are discussed by Arnaud and Kelley (1995), but on the whole, appear to be rare for the Western Alpine domain. The rock units exposed in the Western Alps today seem to be characterised mostly by ages that do not exceed the inferred protolith age of the rock units exposed.

With the new age data presented in this paper we add to the existing picture in that we demonstrate that over a period of several tens of millions of years before the present, the detritus shed by erosion from the Western Alps is characterised by the same set of Cretaceous age groups. This new finding is surprising as it means that at the time of, and very shortly after, the Eocene metamorphism, the detritus is already characterized by these Cretaceous ages. However, the source of the TPB sediments has changed through time and included different Alpine units. Therefore, it is not realistic to think of a uniform reservoir for different sources. Further, the nature of diffusion, governed by transport in terms of exponential transport functions would predict age distributions that essentially would follow a Maxwell-Boltzman distribution (Figure 7). As transport is highly chaotic and dependent on the parameters of individual rock units (e.g. permeability, composition), the actual form of the Boltzman distribution in terms of average, median and skewness will depend critically upon local characteristics of individual rock units. The concentrations of excess argon in the grain boundary reservoir are predicted to be variable in space and time. However, maximum concentrations are unconstrained. Thus, it would be expected that excess argon would produce a range of ages that is constrained on the young side by the actual age of the rock, but on the high age side it is essentially open ended. In fact this is observed in cases where the presence of excess argon is undisputed such as the granulite terrains of Broken Hill (Harrison and McDougall, 1981) and south west Greenland (Pankhurst et al., 1973).



Figure 6. Step heating analyses of Rocchetta and Cassinasco samples.



Figure 7. Maxwell-Boltzman distribution for the gas law (the plot of the number of molecules concentration versus the speed; as the temperature increases, this curve broadens and extends to higher speeds). In addition to temperature under geological conditions permeability and vein density determine the shape of the curve, i.e. maximum concentrations reached form a continuum, and the maximum concentrations, i.e. excess argon ages, have no theoretical limit.

Our data are inconsistent with the excess argon model for pre-Eocene ages as they are showing clearly defined age groups that are essentially constant over extended periods of time. Thus we argue that they should be interpreted to be caused by the effects of inherited argon rather than the effects of excess argon. The subsequent Paleogene metamorphic history of the area (Reddy et al., 2003) has served to obscure the pattern of earlier ages in that in areas of more intense deformation and recrystallisation Eocene and Oligocene ages can be found (see also Scaillet et al., 1992). However, in those areas removed from the zones of intense deformation and recrystallisation, temperature alone during the Eocene and Oligocene was not enough to fully reset the age of existing minerals, and thus pre-Eocene ages survived. This must have been the case for those rocks that eroded mainly during the Miocene in which the effects of Paleogene metamorphism might have been negligible.

CONCLUSIONS

The results of our study strongly suggest that the older, Cretaceous ages found in the Western Alps are not the accidental result of random processes such as the migration of excess argon, but instead a fundamental attribute of the Alpine orogenic belt that can be traced back in time from the Present to at least the early Miocene.

Ages around 107-120 Ma can be interpreted as the result of cooling following UHP metamorphism due to the onset of the Ligure-Piemontese oceanic subduction in the Western Alpine domain. We argue that these ages are consistent with a model that assumes an Early Cretaceous oceanic subduction and subsequent fast cooling/exhumation. The ages around 82-92 Ma could potentially be indicative of the diachronicity of this process, as it probably occurred earlier in the area at present represented by the Sesia Lanzo zone than in the area of the Internal Western Alpine Massifs. Support for this interpretation is that oldest Eo-Alpine ages are recorded at present in the Sesia Lanzo zone (e.g Ruffet et al., 1997; Cortiana et al., 1998; Agard et al., 2002 and references therein). Ages around 68-75 Ma could be indicative of cooling following HP to UHP metamorphism related to the onset of continental collision and subsequent exhumation. Furthermore, a possible mechanism, which would explain the preservation of the HP-UHP metamorphic signals today, could be exhumation of crustal rocks close to the downgoing slab through shear zones parallel to the subducting plate. The subducting plate provides the necessary cooling to counter the radioactive heat production overprinting the thermochronological signal during exhumation (Wijbrans et al., 1993 and references therein).

Recent models on the Western Alpine evolution all dismiss Early Cretaceous ages (e.g. Dal Piaz, 1999; Agard *et al.*, 2002; Reddy *et al.*, 2003) and consider the HP to UHP metamorphism related to the onset of subduction to be no earlier than 70 Ma. On the other hand, evidence of Late Jurassic-Early Cretaceous subduction related to the Tethys, exists in the Eastern Alps and Western Carpathians (Dal Piaz *et al.*, 1995). Therefore, if our interpretation of the new data is correct then detrital 40 Ar/ 39 Ar Cretaceous

ages would be evidence for the relics of a diachronous (earlier in the east than in the west) Tethys subduction. We conclude that the metamorphic history of the Western Alps domain is substantially more complex than current models assume, and the Pre-Eocene tectonic history needs to be seriously re-considered.

ACKNOWLEDGEMENTS

This study has been supported by the Netherlands research centre (NWO). We kindly thank Glen Murrell for his very helpful revision. This paper has benefited from comments of G. Compagnoni, S.P. Kelley, and G.V. Dal Piaz in reaction to the presentation of our research at the 2003 EGS-AGU-EUG conference.

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