Lead isotope systematics of Late Cretaceous - Tertiary Andean arc magmas and associated ores between 8°N and 40°S: evidence for latitudinal mantle heterogeneity beneath the Andes

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Abstract

Lead isotope variability of magmatic arc rocks and associated mineralization of the Central Andes is usually considered to be the result of mixing between a homogeneous mantle and heterogeneous continental crust. About 230 new lead isotope data on the Northern and Central Andes allow us to compare for the first time lead isotope systematics of the Late Cretaceous – Tertiary arc magmatism and associated mineralization along the Andean chain between 8°N and 40°S. Lead isotope compositions indicate mixing between mantle and upper crustal rocks along the whole Andean chain. Additionally, we have found that mantle end-members of the Late Cretaceous – Tertiary magmatism are heterogeneous and systematically shifted towards less radiogenic 206Pb/204Pb compositions from north to south along the Andes. This heterogeneity most likely results from mixing between a low radiogenic mantle, possibly carrying a DMM or EM I component, and a more radiogenic mantle, possibly carrying an HIMU component. Thus, our results imply that lead isotope variability of Andean magmas at the continental scale is caused not only by crustal [...]
Lead isotope systematics of Late Cretaceous – Tertiary Andean arc magmas and associated ores between 8°N and 40°S: evidence for latitudinal mantle heterogeneity beneath the Andes

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ABSTRACT

Lead isotope variability of magmatic arc rocks and associated mineralization of the Central Andes is usually considered to be the result of mixing between a homogeneous mantle and heterogeneous continental crust. About 230 new lead isotope data on the Northern and Central Andes allow us to compare for the first time lead isotope systematics of the Late Cretaceous – Tertiary arc magmatism and associated mineralization along the Andean chain between 8°N and 40°S. Lead isotope compositions indicate mixing between mantle and upper crustal rocks along the whole Andean chain. Additionally, we have found that mantle end-members of the Late Cretaceous – Tertiary magmatism are heterogeneous and systematically shifted towards less radiogenic 206Pb/204Pb compositions from north to south along the Andes. This heterogeneity most likely results from mixing between a low radiogenic mantle, possibly carrying a DMM or EM I component, and a more radiogenic mantle, possibly carrying an HIMU component. Thus, our results imply that lead isotope variability of Andean magmas at the continental scale is caused not only by crustal but also by mantle heterogeneity.

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Introduction

Geochemical data suggest that Andean arc melts are generated in the mantle wedge whence they migrate into and become variably contaminated by continental crust (e.g. Thorpe et al., 1982). This model is supported also by lead isotopes, which, among the radiogenic systems, represent the most extensive database of the Andes, including analyses on both magmatic rocks and associated ore minerals. Indeed, most lead isotope compositions of Central Andean magmatic arc rocks and associated ores define steep linear arrays in conventional isotope plots (Macfarlane, 1995), which are interpreted as mixing lines between a fairly homogenized mantle and upper crustal rocks (Barreiro, 1984).

Many studies have shown that the Pb isotope variability of arc magmas in segments of the Central Andes is mainly due to crustal contamination (e.g. James, 1982; Wörner et al., 1992; Aitcheson et al., 1995). In this paper we discuss 234 new lead isotope data on Late Cretaceous – Tertiary magmatic arc rocks and associated ores of the Northern Andes of Ecuador (N = 165) and mining districts of Peru (N = 69) together with literature data on magmatic arc rocks and associated ore deposits of the Central Andes. Lead isotope compositions of magmatic-related mineralization of the Andes can be used as proxies of magmatic lead compositions because, with few exceptions, they coincide with lead isotope signatures of the associated magmatic rocks (e.g. Puig, 1988; Macfarlane et al., 1990; Chiaradia and Fontbote, 2001). The new data allow us to compare lead isotope reservoirs for arc magmatism of the Northern and Central Andes (8°N–40°S). We present, for the first time, evidence that the sub-Andean mantle is characterized by a systematic decrease in the 206Pb/204Pb ratio from north to south, and propose a possible explanation for this heterogeneity.

The database

This study is based on about 500 lead isotope data, 234 of which are from analyses performed at our laboratories while the remainder are from previous investigations (see database available at http://www.blackwell-sciences.com/products/journals/summat/TER/TER426/TER426sm.htm). They include magmatic arc rocks and related mineralization ( VHMS, porphyry, epithermal, skarn) with Late Cretaceous to Late Tertiary ages. The time span chosen is determined by the homogeneous distribution of magmatism and associated mineralization along the whole Andean chain in the Late Cretaceous – Tertiary, and by the availability of a much larger database than for older periods, thus permitting a reasonable comparison of different segments of the Cordillera.

Ores of province III of Macfarlane et al. (1990) have been excluded because they do not have magmatic signatures, deriving most of the lead from hydrothermal leaching of crustal basements. The Tertiary – Quaternary volcanics of the Arequipa massif and Belén segment in Chile, which have low radiogenic lead isotope signatures due to contamination by lower crust-type basements, have also been excluded.

The Pb isotope data have been subdivided into four latitudinal areas (Fig. 1): 8°N–5°S (Ecuador and Colombia); 5°S–15°S (Northern Peru); 15°S–20°S (Southern Peru); 20°S–40°S (Chile). These subdivisions coincide with major geomorphological features (Huanacamba deflection at 5°S; Arica elbow at 20°S) and/or with lead isotope provinces of Macfarlane et al. (1990) (e.g. the 15°S limit between subprovinces Ib, II to the north and subprovince Ic to the south).
Macfarlane, 1995) we interpret these arrays as mixing lines between the mantle, possibly enriched by pelagic sediments, at the low $^{207}$Pb/$^{204}$Pb (and $^{208}$Pb/$^{204}$Pb) end, and upper crustal rocks at the high $^{207}$Pb/$^{204}$Pb (and $^{208}$Pb/$^{204}$Pb) end. Comparison of the arrays, especially in the uranogenic plot that allows the best discrimination between mantle and upper crust reservoirs, shows three remarkable features (Fig. 2): (1) Arrays are systematically shifted towards lower $^{206}$Pb/$^{204}$Pb values from Ecuador-Colombia to Chile. This supports the conclusions of Macfarlane (1999), who recognized a northward increase in radiogenicity of Tertiary ores of Peru, which, he suggested, could continue into Ecuador and Colombia. (2) The low radiogenic (mantle) end-members of each array contain progressively $^{206}$Pb-poorer lead from north to south. (3) Convergence of the upper ends of the Northern Peru, Southern Peru and Chile trends suggests assimilation of upper crustal rocks with similar Pb isotope compositions in these provinces. This is in part due to the exclusion from our database of magmatic rocks and ores showing the low radiogenic imprint of Proterozoic-type basements. The upper crustal end-member of the Ecuador–Colombia trend has in contrast a higher $^{206}$Pb/$^{204}$Pb ratio suggesting that the crustal basement of this province is isotopically different from that of the Central Andes.

### Results and discussion

Late Cretaceous – Tertiary ores and magmatic arc rocks of each of the four Andean provinces define distinct steep elliptical arrays in conventional plots (Fig. 2), indicating that the majority of lead within each province derives from mixing of two main end-members characterized, respectively, by low and high $^{207}$Pb/$^{204}$Pb and $^{208}$Pb/$^{204}$Pb ratios. The prolateness of the ellipses indicates that the two-end-member mixing is disturbed by secondary sources, by short time-evolved reservoirs, and, probably, by $^{206}$Pb/$^{204}$Pb variability of the main sources. Like previous studies (e.g. Macfarlane, 1995) we interpret these arrays as mixing lines between the mantle, possibly enriched by pelagic sediments, at the low $^{207}$Pb/$^{204}$Pb (and $^{208}$Pb/$^{204}$Pb) end, and upper crustal rocks at the high $^{207}$Pb/$^{204}$Pb (and $^{208}$Pb/$^{204}$Pb) end. Comparison of the arrays, especially in the uranogenic plot that allows the best discrimination between mantle and upper crust reservoirs, shows three remarkable features (Fig. 2): (1) Arrays are systematically shifted towards lower $^{206}$Pb/$^{204}$Pb values from Ecuador-Colombia to Chile. This supports the conclusions of Macfarlane (1999), who recognized a northward increase in radiogenicity of Tertiary ores of Peru, which, he suggested, could continue into Ecuador and Colombia. (2) The low radiogenic (mantle) end-members of each array contain progressively $^{206}$Pb-poorer lead from north to south. (3) Convergence of the upper ends of the Northern Peru, Southern Peru and Chile trends suggests assimilation of upper crustal rocks with similar Pb isotope compositions in these provinces. This is in part due to the exclusion from our database of magmatic rocks and ores showing the low radiogenic imprint of Proterozoic-type basements. The upper crustal end-member of the Ecuador–Colombia trend has in contrast a higher $^{206}$Pb/$^{204}$Pb ratio suggesting that the crustal basement of this province is isotopically different from that of the Central Andes.

### Lead isotope systematics of the Late Cretaceous – Tertiary Andean mantle end-members

An intriguing feature of the Andean lead isotope systematics is the southward decrease of $^{206}$Pb/$^{204}$Pb values of the mantle end-members of the four provinces above defined. The compositional differences of the four mantle end-members cannot be attributed to a single time-evolved reservoir because of the similar Late Cretaceous – Tertiary ages of the investigated magmatic rocks and associated ores in all provinces. We also exclude the possibility that the different mantle end-members are an artefact of crustal contamination. Indeed, arrays of the four provinces being subparallel (Fig. 2), the low radiogenic end-members cannot derive from mixing between a homogeneous low $^{207}$Pb/$^{204}$Pb and $^{208}$Pb/$^{204}$Pb mantle and heterogeneous (variable $^{208}$Pb/$^{204}$Pb) high $^{207}$Pb/$^{204}$Pb and $^{208}$Pb/$^{204}$Pb reservoirs like upper crustal rocks and pelagic sediments. The $^{206}$Pb/$^{204}$Pb spread of the mantle end-members cannot derive either from assimilation of low radiogenic lower crust by mantle-derived melts during melting-assimilation-storage-homogenization (MASH) processes. This would imply enrichment in thorogenic lead concomitant with the southward decrease of $^{206}$Pb/$^{204}$Pb values of the low radiogenic end-members, which is not observed (Fig. 2). In contrast, the isotopic differences of the mantle end-members can be related to mixing between MORB components characterized by different $^{206}$Pb/$^{204}$Pb and consistently low $^{207}$Pb/$^{204}$Pb ($^{208}$Pb/$^{204}$Pb) ratios (Fig. 3). Mixing of DMM and enriched mantle components is considered responsible for MORB isotope variability (Zindler and Hart, 1986). Mixing between a low radiogenic MORB mantle, possibly carrying a DMM or EM I component, and a more radiogenic MORB mantle, possibly carrying an HIMU component, best explains the $^{206}$Pb/$^{204}$Pb range of the Andean mantle end-members (Fig. 3). Note that the mantle end-member of the Ecuador–Colombia trend is $^{206}$Pb richer than E-Pacific MORB (Fig. 3). Strontium and neodymium isotopic constraints on the presence of an HIMU component in the sub-Andean mantle are precluded at the moment by the lack of extensive Sr and Nd data, and, especially, by widespread crustal contamination of Andean magmas. This makes the unidimensional Sr and Nd isotopic systems useless to characterize mantle end-members of the Andean arc magmatism in contrast with the lead isotope system, which allows identification of the mantle end-member by extrapolation of trends in the bidimensional isotope space even in the absence of rocks/minerals directly representing the mantle.

Although the presence of a true HIMU component in the northern sub-Andean mantle remains elusive, mixing of a low radiogenic and a high radiogenic mantle is further revealed in the Northern Andes by Triassic
Fig. 2 $^{208}\text{Pb}/^{204}\text{Pb}$ and $^{207}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ plots of Late Cretaceous – Tertiary magmatic rocks and associated ores of the Andean Cordillera, subdivided according to the geographical provinces of Fig. 1. For the samples analysed at our laboratories, fractions of whole rock and ore mineral lead, previously purified through ion-exchange chromatography on AG1-X8 and AG-MP1 resins, were loaded onto zone refined rhenium filaments using the silica gel technique. Lead isotope ratios were measured on a Finnigan MAT 262 mass spectrometer at the Department of Mineralogy, Geneva (Switzerland), and were corrected for fractionation by a $+0.08\%$ amu correction factor based on more than 100 analyses of the SRM-981 international standard. The analytical uncertainties ($2\sigma$) are $0.05\%$ for $^{206}\text{Pb}/^{204}\text{Pb}$, $0.08\%$ for $^{207}\text{Pb}/^{204}\text{Pb}$ and $0.10\%$ for $^{208}\text{Pb}/^{204}\text{Pb}$. Procedural blanks were always $<120$ pg Pb and insignificant in view of the amounts of lead analysed. The bars on the upper left corner of the diagrams refer to the analytical uncertainties ($\pm 2\sigma$) in our laboratory. The upper crust (UC) and orogen (OR) evolution curves are from Zartman and Doe (1981). The regression lines of each province were calculated on data averaged within $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ frequency intervals of 0.015 and 0.050, respectively, to remove the scatter caused by secondary sources, short time-evolved reservoirs, and $^{206}\text{Pb}/^{204}\text{Pb}$ variability of the two main sources. A simple regression would be strongly biased by the high concentration of data in the central part of each trend due to the fact that the great majority of magmatic rocks of the Andes suffered crustal contamination. Since raw data show that the arrays derive from mixing of two main end-members, the aim of the method chosen was to bias the data by emphasizing these end-members. Nevertheless, frequency intervals that contained less than three analyses were excluded to avoid a bias due to a limited number of data. All linear trends have $R > 0.8$ and $P = 0.0001$ ($P =$ significance level). The numbers refer to the provinces of Fig. 1. $N$ is the number of points on which the regression for each province was based. The lead isotope data used in this study as well as the detailed references used to complement our database can be found at http://www.blackwell-science.com/products/journals/suppmat/TER/TER426/TER426sm.htm.
amphibolites (U–Pb zircon age of 221 ± 17 Ma, Aspden et al., 1995) of the Piedras Group, which is part of a metamorphic complex cropping out in south-western Ecuador. The Piedras amphibolites represent mantle-derived magmas related to the initial intracontinental rifting which led to the separation of the North and South American plates (Litherland et al., 1994). They have MORB-type geochemical features with no signs of crustal contamination (Aspden et al., 1995) and Pb isotope compositions less radiogenic than the E-Pacific MORB field (Fig. 3). Lacking crustal contamination, they allow us to explore Pb isotope geochemistry of the Triassic mantle beneath northern South America. If this mantle had the isotopic composition of the Piedras amphibolites, with a $\mu(238\text{U}/204\text{Pb}) = 9$ and $\alpha(232\text{Th}/204\text{Pb}) = 32$, by Late Cretaceous – Tertiary times can explain the enriched mantle end-member in the Northern Andes (Fig. 3). The possibility that a 206Pb-rich mantle physically different from the Triassic one was the source of the Late Cretaceous – Tertiary magmatism of the Northern Andes (Larson, 1991). These oceanic plateaus, characterized by 206Pb-rich compositions, could have enriched the sub-Andean mantle of the Northern Andes or, if underplated under the continental crust, could have

**What could cause the lead isotope heterogeneity of the Andean mantle?**

At the present stage it is very difficult to identify the cause of the sub-Andean mantle Pb isotope heterogeneity pointed out by our data. As working hypotheses, we propose two tentative scenarios of post-Triassic mixing between a radiogenic mantle, possibly carrying an HIMU component, and a low radiogenic mantle to explain the sub-Andean mantle heterogeneity. Oceanic plateaus generated by the mid-Cretaceous superplume event have been subducted during the Mesozoic beneath Central America and Northern South America but not beneath Central South America (Larson, 1991). These oceanic plateaus, characterized by 206Pb-rich compositions, could have enriched the sub-Andean mantle of the Northern Andes or, if underplated under the continental crust, could have
contaminated mantle-derived magmas on their way to the surface. Another speculative scenario is that Mesozoic plume events occurred at the northern edge of South America, e.g. the 90 Ma old Galapagos hotspot (Sinton et al., 1998) and the 106–82 Ma old hotspot in the Ecuadorian Oriente basin (Barragan and Baby, 1999), could have dispersed $^{206}\text{Pb}$-rich material into the surrounding upper mantle creating the isotopic gradient observed along a distance of several hundred kilometres (see also Schilling et al., 1994; Taylor et al., 1997). Nevertheless, the latter process appears more difficult to reconcile with a subduction environment.

Conclusions

The addition to the existing database on the Central Andes of new extensive Pb isotope data on the Northern and Central Andes has provided for the first time evidence that the Late Cretaceous – Tertiary sub-Andean mantle is systematically $^{206}\text{Pb}$-poorer from north to south along the Cordillera (8°N–40°S). The isotopic gradient of the sub-Andean mantle is best explained by mixing between low radiogenic MORB, possibly carrying a DMM or EM I component, and a more radiogenic MORB, possibly carrying an HIMU component. Our data indicate that mixing of the two mantle components could have occurred between the Triassic and Late Cretaceous. Subduction (and/or underplating) beneath northern South America of $^{206}\text{Pb}$-rich Cretaceous oceanic plateaus or, less likely, diffusion of a Mesozoic plume head in northern South America are speculative processes proposed to explain the enrichment of the northern sub-Andean mantle. Support to these models by Sr and Nd isotopes is at the moment precluded by insufficient data on these isotopic systems, especially concerning arc rocks with basic compositions closely representing mantle signatures.

Our results imply that, although crustal contamination is responsible for most of the Pb isotope variability in segments of the Central Andes (e.g. Davidson and de Silva, 1992; Wörner et al., 1992), mantle heterogeneity is an additional factor responsible for Pb isotope variability of the Andean provinces at the continental scale. They would also document the first example of a possible HIMU-type component in the source of continental arc magmas thus complementing similar conclusions drawn by Hickey-Vargas (1992) on the source of island arc magmatism.

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Supplementary material

The complete data set is available from http://www.blackwell-science.com/products/journals/suppmat/TER/TER426/TER426sm.htm

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