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Timescales and mechanisms of plume–lithosphere interactions: $^{40}Ar/^{39}Ar$ geochronology and geochemistry of alkaline igneous rocks from the Paraná–Etendeka large igneous province

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Abstract

We have determined high-precision $^{40}Ar/^{39}Ar$ ages for alkaline igneous rocks from the western margin of the Early-Cretaceous Paraná–Etendeka large igneous province (Paraguay). These show that small-fraction melt generation occurred beneath the region in two phases; at 145 Ma and 127.5 Ma, i.e. before and at the end of the 139–127.5 Ma Paraná–Etendeka flood-basalt eruptions. Previously published $^{40}Ar/^{39}Ar$ ages for alkaline igneous rocks on the proto-Atlantic coastal margins range from 134 to 128 Ma and indicate that small-fraction melt generation in the east of the province was either synchronous or slightly later than the main pulse of tholeiitic volcanism (between 134 and 132 Ma). Our new $^{40}Ar/^{39}Ar$ phlogopite ages confirm that: (i) the earliest melts associated with the initial impact of the Tristan plume were generated in the west of the Paraná–Etendeka large igneous province and (ii) igneous activity was long lived and immediately predates continental break-up.

The Early-Cretaceous Paraguayan alkaline magmas are silica-undersaturated, enriched in incompatible-trace elements, have very-low initial εNd values and probably represent melts of phlogopite-bearing, carbonate-metasomatised peridotite in the subcontinental lithospheric mantle. Our simple one-dimensional, conductive-heating models suggest that the early-phase (145 Ma) alkaline magmas were emplaced on the margins of the Rio de La Plata craton at the time of sublithospheric impact of the proto-Tristan plume. The late phase (127.5 Ma) of Paraguayan alkaline magmatism is concentrated in an intra-cratonic rift zone and melt generation appears to have been triggered by lithospheric extension, perhaps facilitated by conductive heating and thermal weakening associated with the upwelling Tristan plume.

The location and timing of both alkaline and tholeiitic melt generation in the Paraná–Etendeka province appear to have been significantly influenced by the non-uniform composition and thickness of the South American and south-west African lithosphere. The long duration of Paraná–Etendeka magmatism (17 Myr) relative to other Phanerozoic large igneous provinces (e.g. Siberia, Karoo, and Deccan) may be an artefact of the limited available high-precision age data for CFB-related alkaline igneous rocks.

Keywords: Paraná–Etendeka; $^{40}Ar/^{39}Ar$ geochronology; alkaline igneous rocks; Tristan; mantle plume; large igneous province

1. Introduction

The rates and timescales of emplacement of the vast volumes (1–4 million km^3) of magma associated with

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large igneous provinces (LIPs) provide important constraints on the mechanisms of melt generation. Rapid eruption rates (i.e. over a few million years) reflect adiabatic decompression melting in the convecting mantle whereas a much longer duration of igneous activity (perhaps over 10s of millions of years) is associated with lithospheric melting by heat conduction [1]. Most workers believe that: (i) the presence of anhydrous silicate melts with high-Mg and Ni contents, together with the huge volumes of magma in LIPs, require high mantle potential temperatures ($T_p = \sim 1550$ °C) and (ii) the wide extent of magmatism (over 1000s of km) is caused by lateral spreading of an impacting mantle plume 'starting head'. Many Phanerozoic LIPs are associated with continental break-up (e.g. Deccan, Karoo, Paraná–Etendeka) and it has been proposed that significant lithospheric extension is connected with the main pulse of tholeiitic volcanism [1]. In some cases this rifting did not advance as far as formation of oceanic lithosphere, e.g. Siberia and Columbia River provinces, which alludes to the complex interaction of lithospheric and asthenospheric processes that occur during LIP formation. Alternative models, such as those invoking melting at ambient temperatures ($T_p = \sim 1300$ °C) of a non-peridotite source, i.e. eclogite, cannot adequately explain either the bulk-rock compositions or the spatial and temporal distributions of tholeiitic melts in LIPs such as the Paraná–Etendeka.

In most well-dated LIPs the main pulse of tholeiitic melt generation appears to have occurred in just a few million years (Fig. 1). The Paraná–Etendeka LIP represents one of the world's largest outpourings of basaltic magma ($>1 \times 10^6$ km^3) but, despite the availability of high-precision $^{40}Ar/^{39}Ar$ dating techniques, the duration of tholeiitic volcanism has been the subject of considerable debate [2–6]. This is because tholeiitic melt generation in the Paraná–Etendeka province appears to have occurred over a larger time interval (~ 12 Ma [2]; Fig. 1) than in other large igneous provinces, such as the Deccan (~ 7 Ma [7]), Siberia (~ 7 Ma [8,9]) and Karoo/Ferrar (~ 8 Ma [10–12]). The long duration of tholeiitic magmatism in the Paraná–Etendeka LIP has prompted suggestions that the mechanism of melt generation was different to that of other provinces [13,14]. Nevertheless, fine-grained basaltic rocks are notoriously difficult to date and it is unclear as to whether or not the wide range

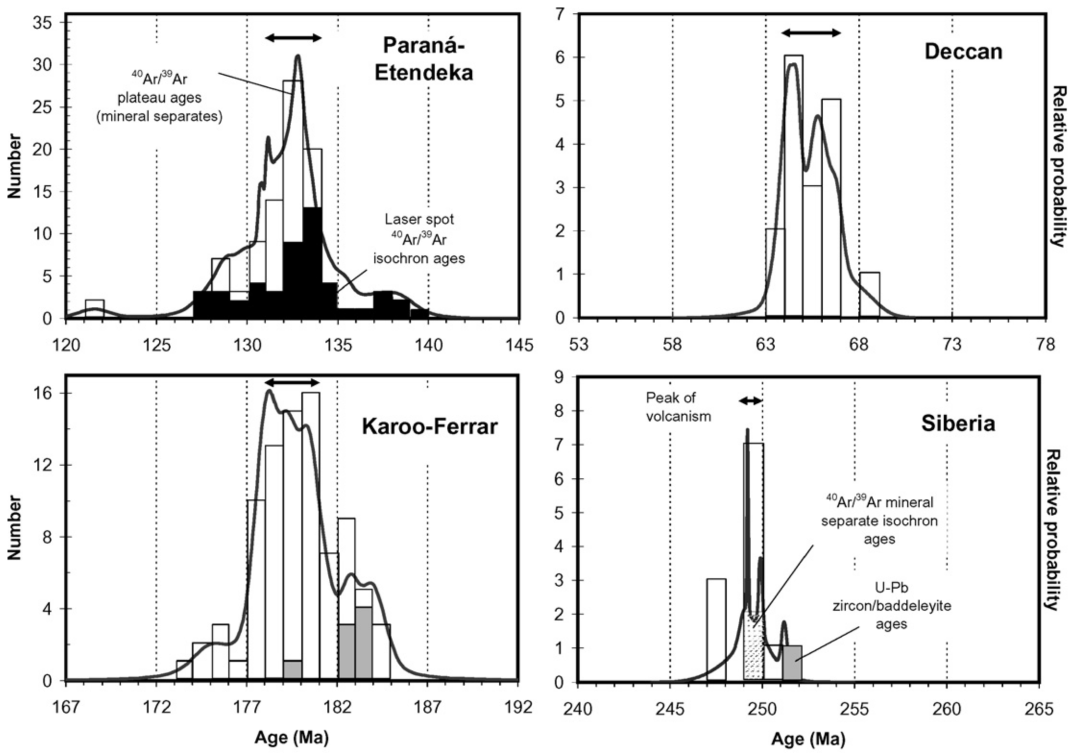

Fig. 1. $^{40}Ar/^{39}Ar$ and U–Pb ages for alkaline and tholeiitic rocks from Phanerozoic large igneous provinces. Published $^{40}Ar/^{39}Ar$ plateau age data have been filtered to remove: (i) whole-rock determinations; (ii) those calculated from steps containing <70% of the total ^{39}Ar released. MSWD values of plotted samples are <2.6 (where given in the literature). All ages have been recalculated to a FCs monitor age of 28.02 [45]. Data sources are given in the Online Background Dataset.

of ages determined for the Paraná–Etendeka province reflects the duration of magmatism or inaccuracies in dating techniques.

In this study, we have focused on the small-volume, mafic, alkaline igneous rocks (lamprophyres, basanites and their fractionated and coarse-grained equivalents) that are temporally and spatially associated with continental flood-basalts (CFBs). The aim of our study is to establish the duration of alkaline igneous activity in the Early-Cretaceous Paraná–Etendeka LIP and, using this geochronological framework, establish a tectonomagmatic model for impacting mantle plume starting heads.

2. Regional setting

The Early-Cretaceous Paraná–Etendeka large igneous province outcrops over an area of 1.2×10^6 km^2 (Fig. 2). Its genesis has been widely linked with the sub-lithospheric impact of the mantle plume now believed to be located beneath Tristan da Cuhna and adjacent ocean islands, in the South Atlantic. The actual role of the mantle plume as a heat and/or a melt source in the genesis of the Paraná–Etendeka magmas has been controversial [1,14–18]. Nevertheless, recent studies have established that (as in other CFB provinces) some tholeiitic melts were generated

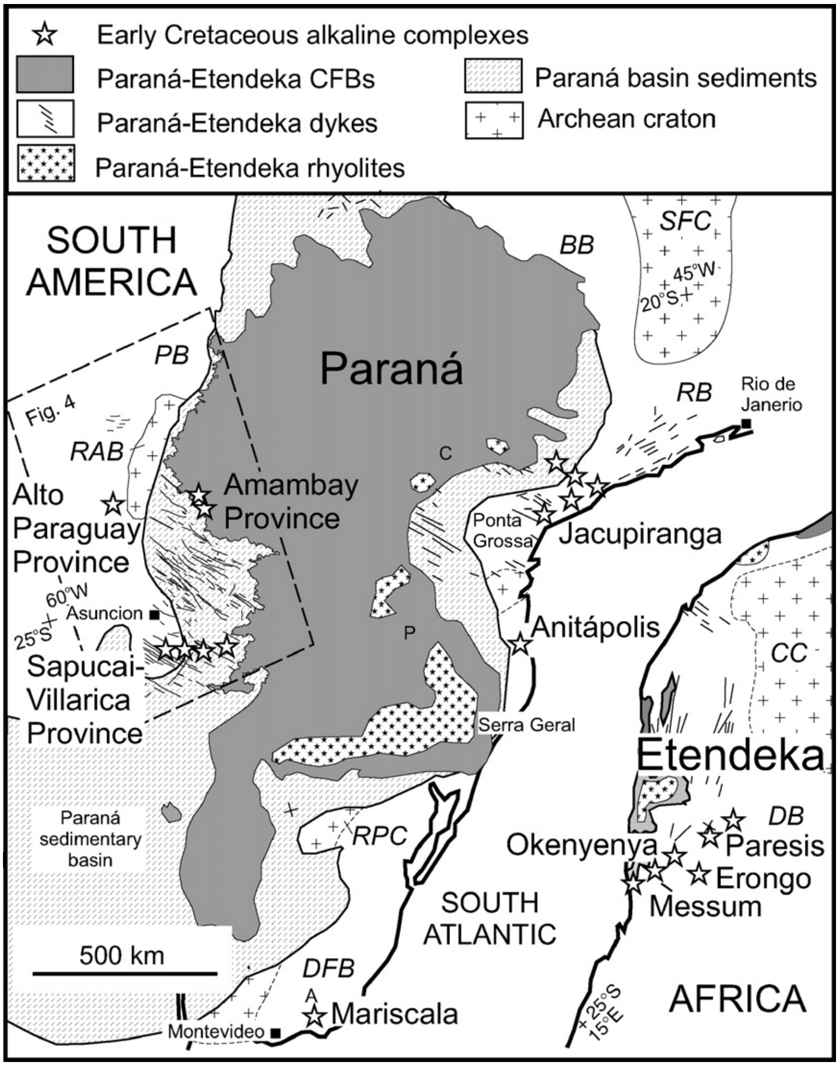

Fig. 2. Distribution of alkaline and tholeiitic igneous rocks in the Paraná–Etendeka large igneous province prior to continental break-up. Modified from Stewart *et al.* [4] to additionally show Early-Cretaceous alkaline igneous rocks [71] and also the approximate locations of cratons and mobile belts [57]. Dyke locations in Paraguay are from [34]. Abbreviations are as follows: BB, Brasilia Belt; DB, Damara Belt; DFB, Dom Feliciano Belt; PB, Paraguai Belt; RB, Ribeira Belt; CC, Congo Craton; LA; Luis Alves Craton; RAB, Rio Apa Block; RPC, Rio de la Plata Craton; SFC, São Francisco Craton; A, Aiüga rhyolites; C, Chapeco rhyolites; P, Palmas rhyolites.

from the convecting mantle although many appear to have been contaminated during ascent through the overlying lithosphere [19]. Small-volume, Early-Cretaceous alkaline magmatism is widespread around the margins of the Paraná–Etendeka province (Fig. 2). Both the tholeiitic basalts and alkaline igneous complexes have an asymmetric distribution about the site of subsequent continental break-up, with a much greater volume (~ 95%) occurring in South America than in south-west Africa (Fig. 2). The cause of this asymmetry has not been established. In contrast, silicic volcanism has a more symmetric distribution about the site of continental break-up which suggests that it is directly linked to lithospheric extension [20].

High-precision $^{40}Ar/^{39}Ar$ ages are available for ~ 100 samples of tholeiitic rocks (flood-basalts and rhyolites [2–6,21–27]) from the Paraná–Etendeka province. Only 6 samples have yielded ages > 136 Ma (Fig. 1) and 4 of these are from the west of the province. All of these 'old' ages were estimated from inverse-isochron plots of $^{40}Ar/^{39}Ar$ laser-spot step-heating analyses of plagioclase grains [2,4] but consistently younger plateau ages (< 132 Ma) have been determined on other samples using furnace step-heating and a defocused $^{40}Ar/^{39}Ar$ laser ([3,5,22,24–27]; Fig. 1). The 'young' $^{40}Ar/^{39}Ar$ plateau ages are for samples collected from the continental margins of South America (Serra Geral escarpment and the Ponta Grossa and coastal dyke swarms [3,25,27] and southern Africa (Etendeka province and Angola [5]; Fig. 2). It is unclear, therefore, as to whether the variations in ages (and hence time-spans of magmatism) are due to differences in analytical techniques and/or sites of sample locations.

Only a few reliable $^{40}Ar/^{39}Ar$ plateau ages have been published for alkaline igneous rocks from the Paraná–Etendeka province and they are mainly for samples from the central (coastal margins of Uruguay and Brazil) and eastern part (Damaraland, Namibia) [4,26,28–31]. These range from 134 to 127.5 Ma (see below) and, together with stratigraphic relationships, suggest that alkaline igneous activity was broadly contemporaneous with or slightly postdates the tholeiitic magmatism. Nevertheless, field relations indicate that alkaline igneous activity in eastern Paraguay (Fig. 3), which is located on the western margin of the Paraná–Etendeka province (Figs. 2 and 4), both pre- and post dates CFB emplacement.

3. Early-Cretaceous alkaline magmatism on the western margin of the Paraná–Etendeka province

Eastern Paraguay consists of Archean and Early–Mid-Proterozoic blocks (belonging to the Rio de la Plata and Amazonas cratons and the Rio Apa block) surrounded by Late Proterozoic mobile belts (Fig. 2). This basement is

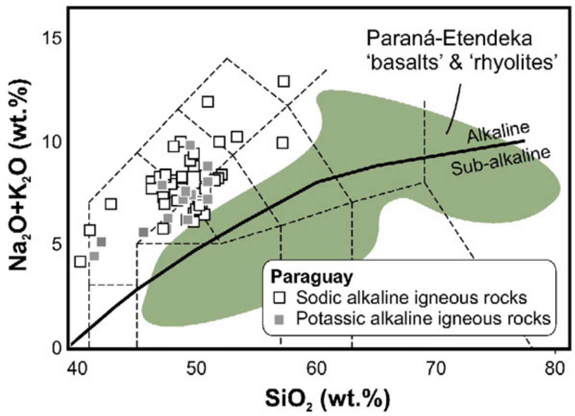

Fig. 3. Total alkalis versus silica plot for Early-Cretaceous magmas from eastern Paraguay. The field of Paraná–Etendeka CFBs and their fractionates is shown for comparison. Solid line shows the divide between alkaline and sub-alkaline igneous rocks after Macdonald and Katsura [72] and the dashed lines show boundaries of IUGS fields after Le Maitre [73]. Data sources are: Online Background Dataset [16,19,63].

overlain by Palaeozoic and Mesozoic Paraná basin sediments and CFB lavas (Fig. 4). There are three known Early-Cretaceous alkaline igneous provinces in eastern Paraguay. The most northerly is the Alto Paraguay *province,* which is located 200 km west of the present-day outcrop of Paraná lavas and on the western margin of the Rio Apa block (Fig. 4). Exposure is poor and the spatial extent of the province unknown; it may overlap with a Permian–Early Triassic alkaline province to the north. Our samples are from thin lamprophyre dykes that intrude Palaeozoic limestones near Vallemi.

The *Amambay alkaline igneous province* is located on the western margin of the Paraná CFB province (Figs. 2 and 4) and outcrops over an area of ~ 1000 km^2. It contains several carbonatite complexes (e.g. Cerro Chiriguelo, Cerro Guazu and Cerro Sarambi [32,33]) that are overlain by flows of tholeiitic basalt. This suggests that the former pre-date CFB emplacement [33]. The nature of the basement beneath the province is unknown. The Amambay province is located north of a large north-west/south-east trending swarm of Paraná dykes (Paranapanema magma type; Background Dataset, [34]). Our samples are mostly from alkaline dykes associated with the carbonatite complexes.

The southernmost of the three Paraguayan alkaline igneous provinces is the *Sapucai–Villarica province* (Fig. 4). This consists of numerous alkaline complexes and plugs and is associated with a 25 to 40 km wide rift structure [35,36] that developed on the north-west margin of Archean basement (probably associated with the Rio de la Plata craton). The rift extends for 200 km from Asuncion to

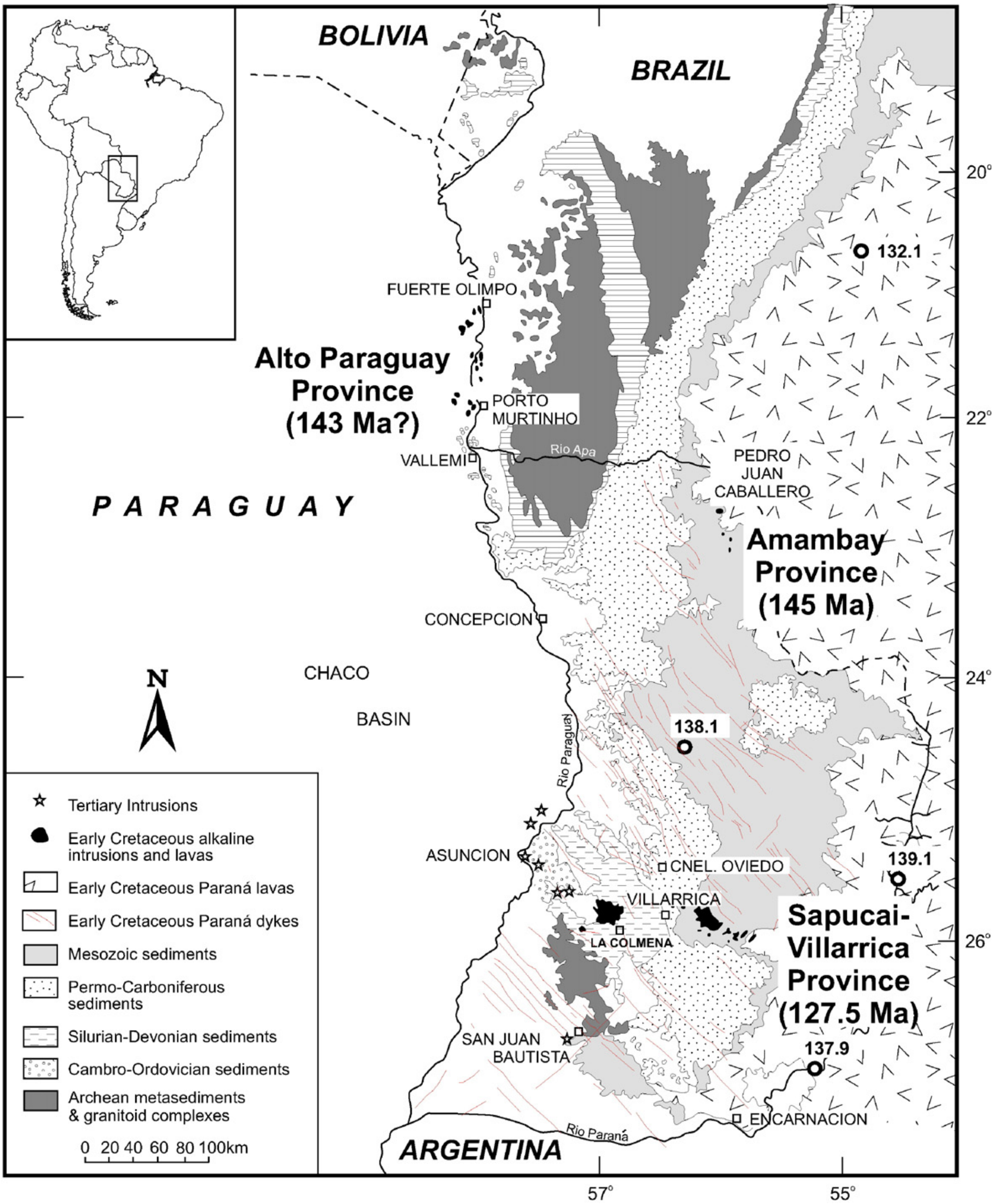

Fig. 4. Location of Cretaceous alkaline igneous provinces in Paraguay. Localities of samples of Paraná dykes previously dated by $^{40}Ar/^{39}Ar$ laser spot step-heating are shown by open circles together with their published ages (in millions of years). $^{40}Ar/^{39}Ar$ age data are from [2,4].

the south east of Villarica (Fig. 4) where it terminates in the Cordillera del Ybytyruzú. Here, alkaline igneous dykes can be seen intruding tholeiitic Paraná basalts.

Fifty-seven samples of sodic (basanites to tephriphonolites) and potassic rocks (lamprophyres) from the three Paraguayan alkaline igneous provinces have been analysed for major, trace and rare-earth elements and a subset for Sr-, Nd- and Pb-isotopic ratios (Online Background Dataset, [37,38]). Seven samples were selected for $^{40}Ar/^{39}Ar$ age determinations.

3.1. Age of Paraná–Etendeka related alkaline magmatism in Paraguay

Conventional K/Ar whole-rock and mineral ages for Paraguayan alkaline igneous rocks range from 66 to 183.5 Ma [39–42]. The large age range reflects loss and excess of ^{40}Ar during weathering and assimilation of mantle-derived ^{40}Ar, respectively. We have eliminated some of these effects by focusing only on ages determined for fresh biotite/phlogopite separates which have >7 wt.% K. This filtered K/Ar dataset suggests that Paraguayan alkaline magmatism occurred between 146.7±9.2 to 124.6±4.2 Ma (Fig. 5).

The $^{40}Ar/^{39}Ar$ step-heating technique permits a separation of the contributions of primary-igneous and secondary-alteration phases to the total Ar and also identification of non-atmospheric Ar. Excess argon is, however, a well-known problem in $^{40}Ar/^{39}Ar$ geochronology and is especially notorious in potassic phases (such as phlogopite). This results in $^{40}Ar/^{39}Ar$ ages that are older than the true closure age and clearly may be significant in the Paraguayan samples. Possible causes of excess argon are:

(i) Pre-emplacement storage of phlogopite. Recent studies have shown that the cores of mantle-derived phlogopite xenocrysts yield ages that are commonly older than the eruption ages of their host rocks. This 'excess' argon is believed to reflect either the age of metasomatism or the storage time in the lithospheric mantle [43]. We have therefore been careful not to include phlogopite xenocrysts in the samples that we have chosen for $^{40}Ar/^{39}Ar$ determinations.

(ii) High solubility of Ar in potassic phases. Argon is strongly incompatible in melts and fluids and is believed to travel along grain boundaries where it may become preferentially concentrated in phlogopite relative to K-poor minerals, such as plagioclase [44].

(iii) Lack of outgassing during cooling. Excess argon is more common in hypabyssal and plutonic rocks than in volcanic systems where it may have been released by outgassing.

3.1.1. $^{40}Ar/^{39}Ar$ methods

$^{40}Ar/^{39}Ar$ age determinations were undertaken on phlogopite separates from 6 lamprophyres and 1 trachyte, collected from the three Early-Cretaceous alkaline igneous provinces in eastern Paraguay (Fig. 4). Samples were selected on the basis of petrographic freshness and because they contained euhedral, phlogopite phenocrysts. Phlogopites were hand picked from crushed material (200–500 μ) and subsequently sealed in air, in quartz vials, and irradiated for 16 h in the CLICIT facility of the Oregon State University (USA) TRIGA reactor. $^{40}Ar/^{39}Ar$ step-heating analyses were performed by M. Pringle at the NERC Argon Isotope Facility at SUERC (Scottish Universities Environmental Research Centre) using a very low-blank, double-vacuum-resistance furnace for the phlogopite step-heating experiments and a CW Nd-YAG laser for the sanidine total-

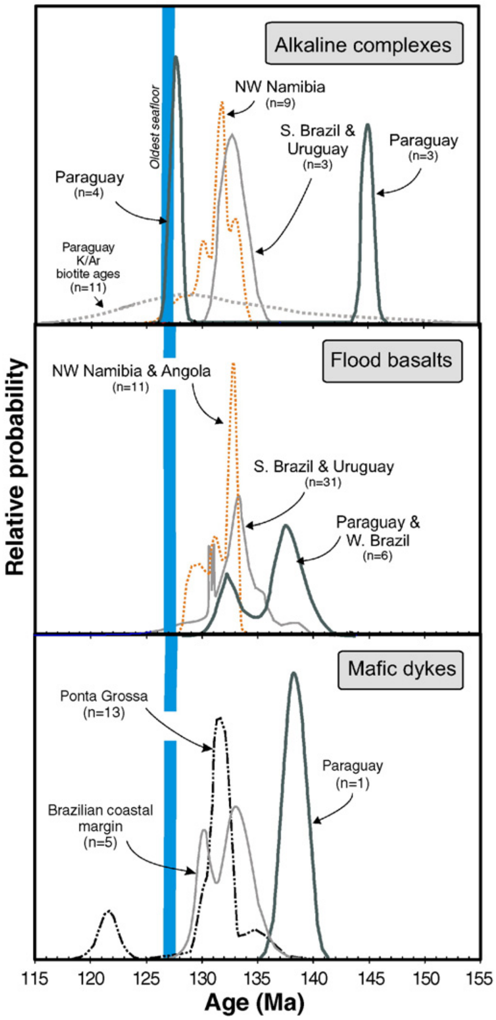

Fig. 5. Age-probability density plots for alkaline and tholeiitic magmas from the Paraná–Etendeka large igneous province. With the exception of K/Ar biotite/phlogopite ages for Paraguayan alkaline complexes, all other ages are $^{40}Ar/^{39}Ar$ and were determined by furnace step-heating of plagioclase or biotite/phlogopite grains or laser-spot step-heating and have been corrected to FCs=28.02 Ma [45]. Published age data have been filtered to remove: (i) plateau ages calculated from steps containing <70% of the total ^{39}Ar released and (ii) ages with MSWD>2.6. The age of the oldest seafloor at this latitude (127 Ma) is shown for reference [49]. K/Ar ages are for biotite/phlogopite separates containing >7 wt.% K [32,33,39,41]. $^{40}Ar/^{39}Ar$ phlogopite plateau ages for Paraguayan rocks are from Table 1. Additional data are from [2–6,22,24–28,30,31,46,48,74].

fusion analysis. The neutron-fluence parameter, J, varied by 0.3%. The neutron flux was monitored using Fish Canyon Tuff sanidine (FCs) at 27·62 Ma. The age of this standard has, however, been revised to 28.02±0.16 [45] and we have followed Hawkesworth *et al.* [13] and recalculated both our ages and previously reported $^{40}Ar/^{39}Ar$ ages for the Paraná–Etendeka province relative to this (Online Background Dataset). The revised age for FCs produces a 1.4% difference in our calculated $^{40}Ar/^{39}Ar$ ages and 0.6% difference for previously published Paraná–Etendeka ages.

Apparent ages were calculated using both $^{40}Ar/^{39}Ar$ age plateau spectra and isochron analyses of phlogopite separates (Supplementary Figure). Acceptable plateau ages are defined by the release of >70% of the total ^{39}Ar gas, in five or more successive steps, concordant within 2σ error and showing no resolvable slope. Ages with a mean square weighted deviate (MSWD) of <2.6 were considered to be the most reliable. Errors are reported as 2σ of analytical precision. Plateau age uncertainties do not include uncertainties in the age of FCs. Excess ^{40}Ar may produce false plateau ages that are older than the actual cooling age and, in order to counteract this problem, we have also plotted data for all samples (excluding the discordant steps from the plateau diagrams) on isotope correlation diagrams (Supplementary Figure). Calculated inverse isochron ages are in general unaffected by excess ^{40}Ar and, in all cases, the inverse isochron ages are indistinguishable from the plateau ages at the 95% confidence level. Each sample gave a well-defined isochron with a $^{40}Ar/^{36}Ar$ intercept close to the atmospheric ratio of 295.5.

3.1.2. Results

All of our samples produced stratigraphically consistent ages but we have identified excess argon in one of these. The argon release spectra of our only sample from the Alto Paraguay alkaline igneous province (93SOB115, Supplementary Figure) shows high apparent ages at low temperatures and the 'plateau' is defined by only 3 steps which represent 70.5% of the cumulative ^{39}Ar released (Supplementary Figure). The plateau age of 158.67±2.07 has a very high MSWD (17.62, Table 1) and we do not consider this to represent a reliable crystallisation age for the rock. This $^{40}Ar/^{39}Ar$ age is significantly older than those determined for other Paraguayan alkaline igneous rocks and is also older than previously determined conventional K/Ar phlogopite ages for the Vallemi dykes of 143±2 Ma (Gibson, Thompson, Mitchell unpubl. data) and 137.4±5.4 Ma [32].

All of the other samples produced relatively undisturbed spectra and yielded well-defined plateaux with >75% of the total ^{39}Ar released (Supplementary Figure). Two dykes from the Amambay alkaline igneous province (93SOB124 and 143) gave reliable $^{40}Ar/^{39}Ar$ plateau ages of 144.99±0.89 Ma and 144.95±0.93 Ma. The weighted average age for the Amambay alkaline igneous province of 144.97±0.63 Ma is within the range of conventional K/Ar dates for fresh biotite/phlogopite separates from Cerro Chiriguelo, Cerro Sarambi and Arroyo Gasory (135±7 Ma to 147±

Table 1
$^{40}Ar/^{39}Ar$ ages determined on phlogopite separates from Paraguayan alkaline igneous rocks

Locality	Sample no.	Rock type	Plateau age (Ma, ±2σ)	MSWD	No of steps	Total ^{39}Ar released (%)	Isochron age (Ma, ±2σ)	J value	Plateau age recalculated to FCs=28.02 Ma (Ma)
Alto Paraguay									
Valle Mi	93SOB115	Lamprophyre	156.40±2.07	17.62	3	70.5	155.75±4.39	0.005097	158.67
Amambay									
Arroyo Gasory	93SOB124	Trachyte	142.92±0.89	1.7	8	85.8	141.07±2.3	0.005086	**144.99**
Cerro Sarambi	93SOB143a	Lamprophyre	142.88±0.93	1.14	7	87.2	141.90±1.2	0.011115	**144.95**
Cerro Sarambi	93SOB143b	Lamprophyre	142.89±0.93	1.6	7	73.5	142.00±2.2	0.011284	**144.96**
Sapucai Villarica									
Sotu–Rugua	93SOB3a	Lamprophyre	126.08±0.79	0.41	8	88.1	126.19±0.83	0.011306	**127.91**
Sotu–Rugua	93SOB3b	Lamprophyre	125.84±0.79	0.64	9	100	125.81±0.83	0.011147	**127.66**
Canada 1	93SOB46	Lamprophyre	125.31±0.78	2.6	6	84.9	125.70±1.8	0.005114	**127.12**
Tacuarita	93SOB63	Lamprophyre	128.23±1.40	18.0	8	75.3	128.80±1.90	0.005103	130.09
Catalan	93SOB106	Lamprophyre	127.60±1.10	4.5	9	61.1	127.50±1.20	0.005109	129.45

Full analytical results are given in the Online Background Dataset.
Bold font denotes preferred ages.
Plateau age includes 2σ j-error of 0.6%.

9 Ma [32,33,41]) but error bars for the $^{40}Ar/^{39}Ar$ ages are significantly less.

Age estimates for only three of the five phlogopite separates from the Sapucai–Villarica province had MSWD values < 2.6 and these have $^{40}Ar/^{39}Ar$ plateau consisting of > 75% of the total ^{39}Ar released. Phlogopite separates from the Sotu–Ruguá (Cerro Santa Tomas) dyke (93SOB3) yielded $^{40}Ar/^{39}Ar$ plateau ages of 127.91 ± 0.79 Ma (MSWD = 0.69) and 127.66 ± 0.79 Ma (MSWD = 0.41) and phlogopite from Canada 1 (93SOB46) gave a $^{40}Ar/^{39}Ar$ plateau age of 127.12 ± 0.78 Ma (MSWD = 2.61). The weighted mean of these three ages is 127.56 ± 0.45 Ma. Sample 93SOB63 and 106 have 'humped' spectra (Supplementary Figure), showing Ar loss at low temperature and Ar recoil effects at high temperatures. These yielded plateau ages of 130.09 ± 1.40 Ma and 129.45 ± 0.79 Ma with MSWDs of 18.0 and 4.5, respectively. Previous conventional K/Ar ages for biotite/phlogopite separates (with K > 7 wt.%) from the Sapucai–Villarica province ranged from 124.6 ± 8.4 Ma to 132.9 ± 10.10 Ma. A similar $^{40}Ar/^{39}Ar$ plateau age of 127.56 Ma has also been quoted for Cerro Santa Tomas ([26] recalculated to FCs = 28.02).

4. Comparison with previously determined ^{40}Ar/^{39}Ar ages for Paraná–Etendeka magmas

Our new $^{40}Ar/^{39}Ar$ ages suggest that Early-Cretaceous alkaline igneous activity in the Paraná–Etendeka province occurred over a longer period of time (17 Myr) than has previously been established from published high-precision $^{40}Ar/^{39}Ar$ and U–Pb ages.

4.1. Alkaline igneous rocks

Whole-rock $^{40}Ar/^{39}Ar$ and U–Pb age determinations are available for Early-Cretaceous alkaline igneous intrusive complexes from Damaraland (north-west Namibia, Fig. 2). Contact relations indicate that, on the eastern fringe of the Paraná–Etendeka province, most of the alkaline intrusives post-date the CFB lavas. A $^{40}Ar/^{39}Ar$ whole-rock plateau age of 138.00 ± 0.8 Ma (MSWD = 0.73) has been reported for a comendite flow from Paresis ([31]) but this age is stratigraphically inconsistent with the $^{40}Ar/^{39}Ar$ K-feldspar plateau age of 131.44 ± 0.6 Ma (MSWD = 1.16) determined for an underlying quartz–feldspar-porphyry flow [46]. Furthermore, the Paresis alkaline complex post-dates the eruption of the Etendeka flood basalts which are believed to have been emplaced < 134 Ma (see below). Field relationships indicate that the alkaline plugs of the Erongo complex (Fig. 2) also postdate the Etendeka flood basalts; a biotite separate from one of these gave an $^{40}Ar/^{39}Ar$ plateau ages of 131.48 ± 2 Ma [28]. The youngest published $^{40}Ar/^{39}Ar$ age for CFB-related alkaline magmatism in north-west Namibia is for the Messum complex. Biotite separates from late syenites gave an $^{40}Ar/^{39}Ar$ isochron age of 127.86 ± 2.4 Ma. An Rb–Sr age of 126.6 Ma has been determined for the Okenyenya complex but this has a 2σ error of ± 7.3 Ma [31,47].

$^{40}Ar/^{39}Ar$ ages for alkaline magmatism on the southern Brazilian and Uruguayan coastal margins (e.g. Jacupiranga, Anitapolis, Mariscala, Fig. 2) range from 133.7 to 131.8 Ma [4,26,31,48]. These complexes are similar in age to the spatially associated tholeiitic flows (Fig. 5a) but are distinct from a later phase of alkaline magmatism.

4.2. Continental-flood basalts and rhyolites

The first phase of alkaline magmatism in the Paraná–Etendeka province pre-dates the earliest tholeiitic magmatism by ~ 6 Ma. $^{40}Ar/^{39}Ar$ ages suggest that the flood basalts were erupted over a 12 Ma time interval, between 139.1 and 127.5 Ma, with the main pulse of tholeiitic magmatism between approximately 134 and 132 Ma (Fig. 5b [2–4,23,25]). Importantly, the oldest ages available for both the alkaline and tholeiitic volcanism are almost all from the west of the province (Fig. 6). The youngest phase of alkaline magmatism in eastern Paraguay was approximately contemporaneous with the emplacement of the youngest flood basalts, which occur on or near the Brazilian and Uruguayan coastal margins and the end of silicic volcanism at 127.7 Ma [2,4–6,27,28].

$^{40}Ar/^{39}Ar$ age determinations for tholeiites from the NW/SE trending Ponta Grossa dyke swarm (Fig. 2) and the coast-parallel dyke swarm of south-east Brazil suggest that major lithospheric extension occurred between 135 Ma and 130 Ma but may have continued to 121.5 ± 1.3 Ma [2,25,27,48]. The onset of dyke emplacement predates the formation of the oldest South Atlantic sea floor (Chron 4, 127 Ma [49]) at this latitude by ~ 8 Ma (Fig. 5c). A NW/SE trending tholeiitic dyke from the eastern Paraguay swarm gave an $^{40}Ar/^{39}Ar$ age of 138.1 ± 2 Ma [4], which suggests that the onset of lithospheric extension may, in fact, have occurred in the western part of the CFB province.

5. Melt generation processes associated with the sub-lithospheric impact of the proto-Tristan mantle plume starting-head

The interpretation of the melt generation processes linked to the impact of subcontinental mantle plumes has been controversial. Detailed geochronology and geochemistry for the Paraná–Etendeka province combined

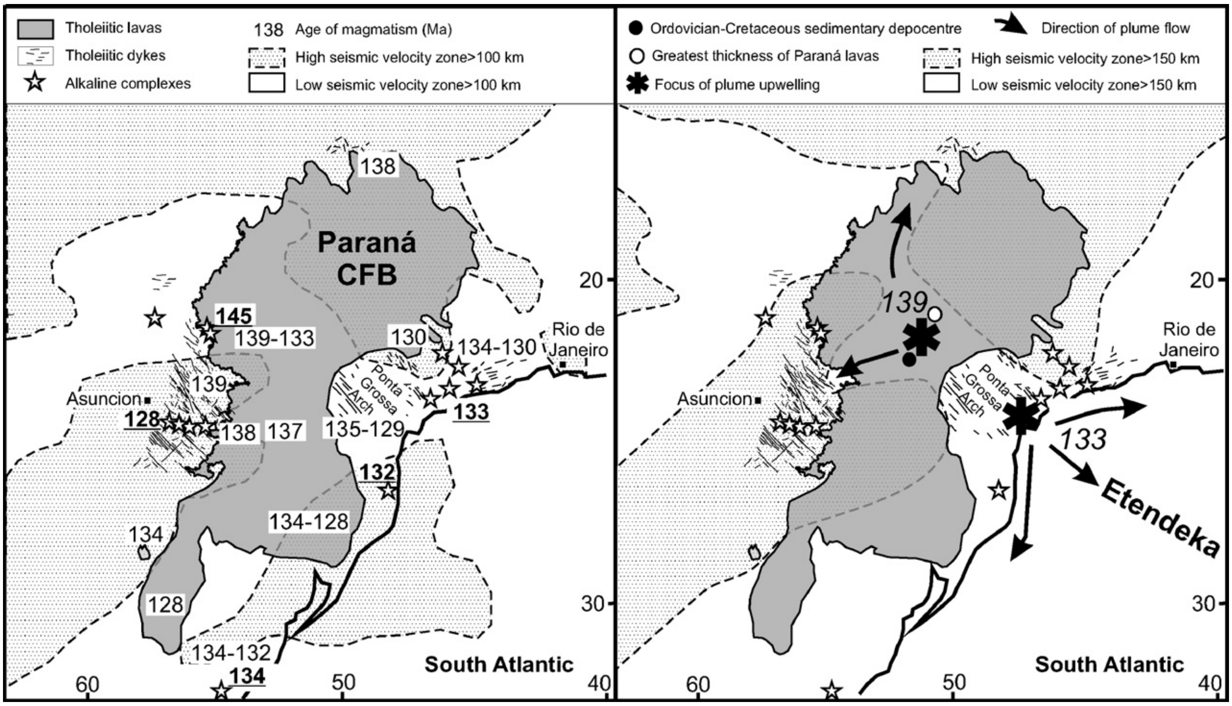

Fig. 6. (a) Spatial variation of high-precision $^{40}Ar/^{39}Ar$ ages determined for alkaline (shown in bold font and underlined) and tholeiitic rocks from the Paraná–Etendeka large igneous province [2–6,21–24,26–28,30,31,48,74]. Also shown are regions of high- and low-seismic velocity at depths > 100 km [50]. (b) Locations of maximum plume upwelling at 139 Ma and 133 Ma have been estimated on the basis of: spatial variations in $^{40}Ar/^{39}Ar$ ages, regions of low-seismic velocity at depths > 150 km [50]; location of the depocentre of Ordovician–Cretaceous sediments and the greatest accumulation of Paraná lavas [75]; and reconstructions of plate motions between 140 and 130 Ma [61].

with the results of seismic–tomographic experiments have, however, allowed us to constrain the mechanisms of melt generation more precisely than has been possible in previous studies.

5.1. Lithospheric thickness

A critical factor in understanding melt-generation processes is the thickness of the mechanical boundary layer (MBL), i.e. the rigid part of the lithosphere where heat is transferred by conduction and long-term geochemical anomalies can develop. The MBL is shallower than the thermal (seismic) lithosphere and represents the minimum depth of asthenospheric upwelling. For example, adiabatic decompression melting in an upwelling mantle plume of fertile peridotite, with a potential temperature (T_p) of 1550 °C, will only occur if the MBL is <165 km thick [1]. Below ~ 125 km, the degree of isentropic melting will be small (<~ 5%) and anhydrous mafic alkali melts form. At shallower depths the amount of melting will increase dramatically (to ~ 20% at 70 km) and melts will have a tholeiitic composition [1].

Heterogeneity in thickness of the lithosphere may have been an important influence on the location, duration and style of mantle melting associated with the impacting proto-Tristan plume starting-head. The results of a recent seismic tomographic study of South America show that the Paraná basin has a non-uniform *S*-velocity structure which, allowing for the lateral resolution of the study, correlates with previously identified tectonic units [50]. At depths > 150 km, high velocities are present beneath the major cratons and low velocities are associated with some of the adjacent mobile belts. Low velocities observed in the Proterozoic mobile belts at depths typically >100 km are probably due to higher geothermal gradients (i.e. thinner lithosphere) and a more fertile mantle. An important feature of the *S*-velocity map of van der Lee *et al.* [50] is the area of low velocity (below 100 km depth) beneath the central Paraná basin (Fig. 6); this correlates with the location of thickest accumulation of sediments (5 km) and CFB lavas (1.7 km), and also a gravity high [51,52]. The sediments range from Ordovician to Cretaceous in age, which indicates that relatively thin lithosphere was present beneath the central Paraná basin prior to the impact of the proto-Tristan plume.

The region of low seismic velocity at depths > 100 km extends south-east from the depocentre, to beneath the

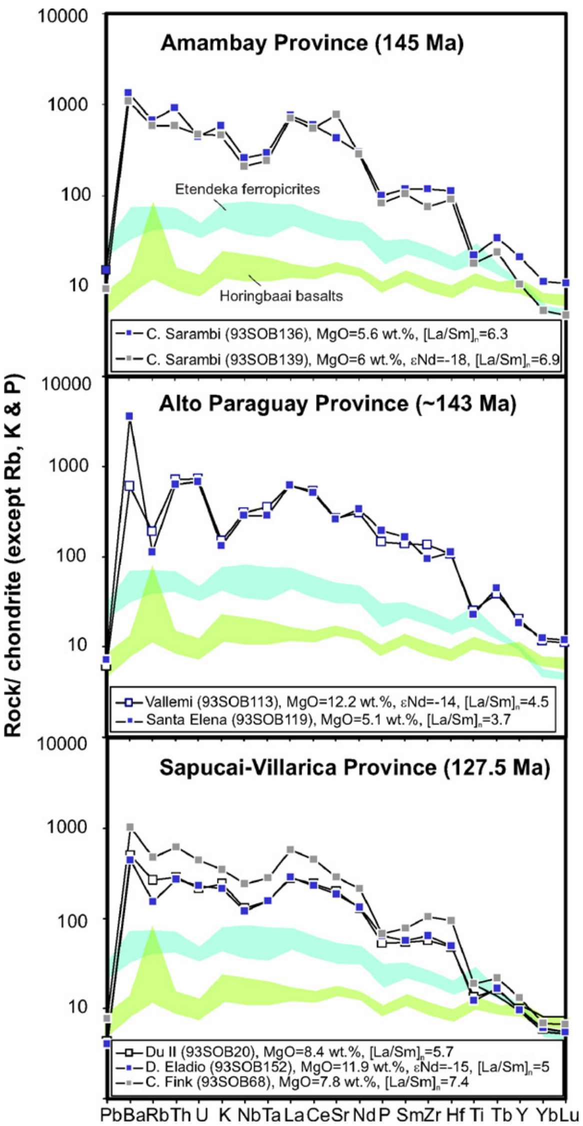

Fig. 7. Primitive-mantle-normalised multi-element plots [76] for Early-Cretaceous mafic sodic (open symbols) and potassic (closed symbols) alkaline igneous rocks from the western margin of the Paraná–Etendeka large igneous province. Fields for uncontaminated melts (Horingbaai dykes and Etendeka ferropicrites) from the Tristan plume starting head are shown for comparison. Data are from Online Background Dataset, [19,62].

Ponta Grossa Arch (Fig. 6). The latter is a north-west-trending tectonic feature exposed on the eastern margin of the Paraná Basin that is partially covered by sediments and lavas but is believed to have been active since the Palaeozoic. It is associated with a major swarm of tholeiitic dykes that fed the Paraná CFB. Tholeiitic dyke swarms along the Rio coast and in north-eastern Paraguay also correlate with regions that have low-velocity below 100 km. We note that the tholeiitic dykes associated with the Sapucai–Villarica rift in southern Paraguay do not coincide with a low seismic-velocity zone. This may be due to the resolution of the seismic tomographic study. Most of the Early-Cretaceous alkaline igneous rocks in South America occur on the margins of regions with high seismic-velocity at depths > 150 km. An exception is Anitápolis, in southern Brazil, which is in a region where there was very low resolution.

The apparent correlation of low seismic velocity regions with Paraná volcanism and the Palaeozoic–Mesozoic sedimentary depocentre indicates that these zones of lithospheric extension and mantle upwelling have not re-thickened conductively to that of the surrounding cratonic lithosphere during the last 130 Myr. Sub-cratonic lithospheric mantle is only gravitationally stable at thicknesses of > 150 km because it is composed of Mg-rich peridotite (i.e. harzburgite) whereas the equilibrium thickness of more fertile mantle, that would result from Phanerozoic re-thickening at the base of the MBL, would be ~ 100 km [53]. Some of the low seismic velocity regions beneath the Paraná and adjacent areas [50] may therefore be due to present-day, incipient partial melting of the convecting mantle beneath regions that had previously undergone Phanerozoic lithospheric thinning.

5.2. Melt source regions

The comparable incompatible-trace-element and isotopic signatures of the early- and late-phase mafic alkaline igneous rocks from the western margin of the Paraná–Etendeka CFB province (Figs. 7 and 8) suggest that they

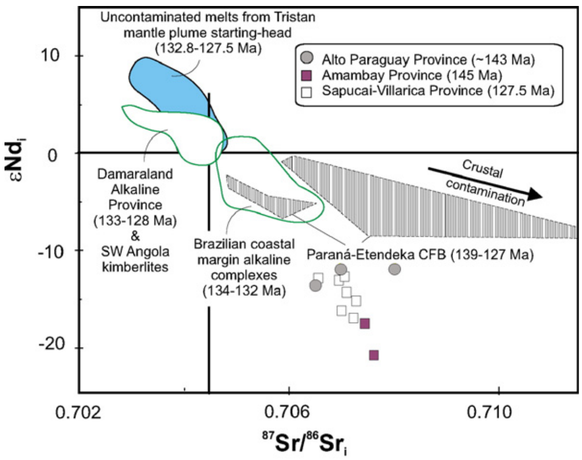

Fig. 8. Variation of initial $^{87}Sr/^{86}Sr$ with initial εNd values for Early-Cretaceous mafic alkaline igneous rocks from the Paraná–Etendeka large igneous province. Fields for uncontaminated melts from the Tristan plume starting head (Horingbaai dykes and Etendeka ferropicrites) and Paraná–Etendeka CFB are shown for comparison. Data are from: [17–19,32,37,62,63,77–81].

were derived from similar mantle source regions. High concentrations of large-ion-lithophile elements, enriched incompatible-trace-element signatures (e.g. $[La/Yb]_n = 25–125$) and relative depletions in high-field-strength elements (e.g. $[La/Nb]_n$ ratios = 1.5–3.5) suggest that the Paraguayan alkaline igneous rocks were formed by melting of a previously metasomatised source. Their low εNd values (−11.8 to −21.6) indicate that enrichment of the Paraguayan lithosphere must have occurred in the Proterozoic and that the MBL was the predominant melt source region [16,54]. The silica-undersaturated nature of the sodic and potassic alkaline igneous rocks, and the high Ni contents (up to 325 ppm) of the most mafic samples, indicate that they may have been generated from a carbonate-metasomatised peridotite source.

The combined high Ba (up to 8600 ppm) and K_2O contents (up to 8.5 wt.%) of the mafic alkaline melts suggest that melting of phlogopite rather than amphibole was the dominant source of K; mineral–melt partition coefficients (*D* values) for both K and Ba are much higher for phlogopite than amphibole, e.g. [55]. Melting of phlogopite also produces silica-undersaturated melts with high K/Na ratios whereas amphibole-derived melts are more silica-saturated. Additionally, phlogopite has $D_{Rb} > D_{Sr}$ and melting of this phase in the source would explain the relatively high initial $^{87}Sr/^{86}Sr$ ratios (0.707–0.708 [16,54]) of the alkaline melts (Fig. 8). The negative HFSE (i.e. Nb, Ta and Ti) anomalies in the Paraguayan alkaline igneous rocks (Fig. 7) may be due to: (i) the relatively high *D* values for these elements between clinopyroxene and carbonate melts; (ii) chromatographic effects during the ascent and crystallisation of metasomatic melts, such that the early precipitation of HFSE-rich minerals causes a relative depletion of these elements in subsequently formed phases; or (iii) metasomatic enrichment by subduction-related silicate melts.

For other Paraná–Etendeka alkaline provinces mantle source enrichment appears to have been more recent and of a different style. For example, mafic alkaline igneous rocks from the Brazilian coastal margin and Namibia (Fig. 2), are characterised by higher concentrations of HFSE (e.g. $[La/Nb]_n = 0.38–2.2$) and εNd values (4 to −7, Fig. 8 [17,19]).

5.3. Conductive heating and extension of the lithosphere

We have constructed a simple, one-dimensional, time-dependent, conductive-heating model to constrain the genesis of the small-volume volatile-rich alkaline igneous rocks on the western margin of the Paraná–Etendeka province (Fig. 9). The initial steady-state temperature for the crust, mechanical- and thermal-boundary layers were calculated using equations and parameters from McKenzie *et al.* [56]. The thickness of the crust beneath eastern Paraguay was assumed to be 40 km [57] and the surface temperature 0 °C. In our models, we have assumed that both the sodic and potassic melts are derived from phlogopite-rich veins residing in the SCLM beneath the cratonic margins (see above). It can be seen from Fig. 9 that the slope of the

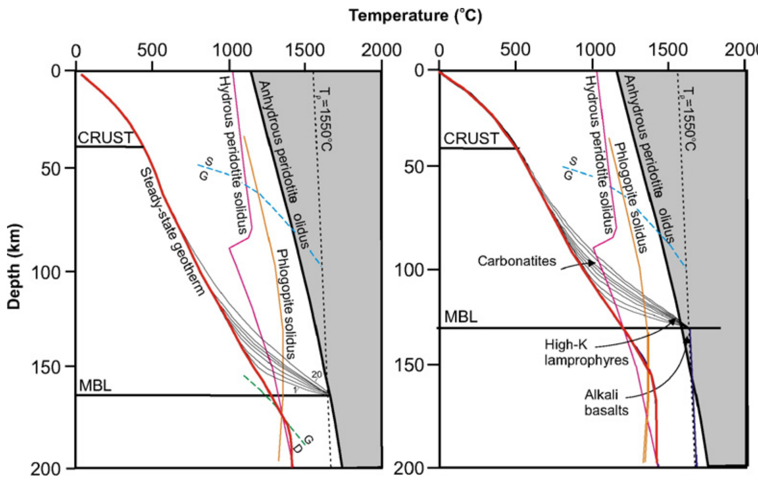

Fig. 9. Conductive-heating model for alkaline igneous rocks on the western margin of the Paraná–Etendeka large igneous province. The parental melts are believed to have been derived from phlogopite-bearing peridotite in the mechanical boundary layer (MBL, see text for discussion). The initial (i.e. steady-state) geotherm is calculated for a mantle potential temperature of 1300 °C, a MBL thickness of (a) 165 km and (b) 130 km, and a crustal thickness of 40 km using the method of McKenzie *et al.* [56]. The change in temperature 1, 2, 4, 6, 8, 10 and 20 Myr after the temperature at the base of the MBL was increased by 250 °C is also illustrated. Solidus curves for anhydrous peridotite, hydrous peridotite and phlogopite are from [58,82,83]. The locations of the garnet–spinel (GS, [84]) and graphite–diamond (GD, [85]) phase boundaries are shown for reference.

phlogopite solidus [58] is similar to the mantle adiabat (0.6 °C km^{-1}) and for a thick MBL, thinning by a β factor of >1.3 would be required to cause melting of metasomatic veins without any increase in temperature. There is, however, no evidence of uplift and/or lithospheric thinning prior to emplacement of the earliest phase of alkaline magmatism on the western margin of the Paraná–Etendeka.

The increase in temperature with depth above the base of the MBL was calculated at different time increments, between 1 and 20 Ma, using the conductive heating equations of Carslaw & Jaeger [59]. In our model, the initial basal temperature was increased instantaneously by 250 °C to represent the temperature difference between ambient mantle (T_p=1300 °C) and the plume starting head (T_p=∼ 1550 °C), where the base of the MBL and TBL are at 130 and 160 km, respectively. Our model differs from previous ones for the Paraná–Etendeka province (e.g. [14]) in that we have assumed: (i) lithospheric mantle melting involves phlogopite-rich veins (see above) rather than wholesale melting of hydrated peridotite; (ii) the lithosphere had a non-uniform thickness and composition at the time of plume impact.

Fig. 9 shows that at the base of 130 km thick MBL, the steady-state geotherm is ∼ 150 °C below the phlogopite solidus. Conductive heating resulting from an increase in temperature of 250 °C at the base of the MBL will raise the temperature of the lithosphere over a depth of ∼ 70 km in a 20 Myr time interval. The most dramatic increase in temperature takes place at the base of the MBL and our model shows that melting of phlogopite-rich veins would occur almost immediately following plume impact. Certainly, within a few million years, conductive heating could generate melt from a 10 km thick metasomatised layer at the base of the MBL. At these temperatures and pressures, only small degrees of partial melting would occur within the upwelling plume. This may explain the presence of small volumes of fractionated alkali basalts (i.e. trachytes) in eastern Paraguay at 145 Ma.

The large time interval between the 2 phases of Paraguayan alkaline igneous activity suggests that conductive heating at the base of the lithosphere alone was not enough to cause the genesis of small-fraction melts in the 127.5 Ma Sapucai–Villarica province. This youngest and most voluminous phase of Early-Cretaceous alkaline magmatism in Paraguay is located at a greater distance from the margin of the Rio de la Plata craton than the early-phase magmatism (Fig. 6) but is concentrated in a well-defined rift zone and local lithospheric thinning may have triggered melting of the metasomatised lithosphere.

5.4. An integrated plume impact model for the Paraná–Etendeka large igneous province.

Asthenospheric upwelling associated with the ascent of the Tristan plume starting-head would have been focused on (i) pre-existing thin spots, such as that beneath the central part of the Paraná basin (Fig. 6), and (ii) weak zones at the margins of cratons and mobile belts [60]. These would have been regions of maximum uplift and extension at the time of plume impact and may also have been sites of initial melt generation in the convecting mantle. Our simple thermal model, integrated with some of the findings of previous geochronological, geochemical and geophysical studies, suggests that the following sequence of events occurred in the genesis of the Paraná–Etendeka LIP:

(i) *Localised melting of metasomatised lithosphere by conductive heating associated with the impact of the Tristan plume on the base of the sub-cratonic lithosphere.* This appears to have rapidly mobilised and exhausted (∼ 1 Myr) readily-fusible, phlogopite-rich veins at the base of the MBL beneath the cratonic margins in the west of the Paraná–Etendeka province.

(ii) *Lithospheric extension focused on a pre-existing region of thin lithosphere, 6 Myr after plume impact*. The *S*-velocity maps of van der Lee *et al.* [50] suggest that such a 'thinspot' is located beneath the central part of the Paraná basin, close to the postulated site of the plume axis (Fig. 6 [61]). Once the MBL had thinned to <70 km, tholeiitic magmas were generated in the underlying upwelling convecting mantle and transported laterally within the crust in large dyke swarms, e.g. eastern Paraguay, to their subsequent eruption sites in the western and northern parts of the province. This contrasts with Hawkesworth *et al.* [13] who have proposed that the tholeiitic melts generated in the first 10 Myr after plume impact, were derived by conductive heating of thick (∼ 150 km) hydrous lithosphere and only subsequently were melts generated from the convecting mantle.

(iii) *Major lithospheric extension and rapid melt production 10 Myr after plume impact*. This occurred in the Ponta Grossa Arch and along the site of the proto coastal margin (135–126 Ma). White and McKenzie [1] argued that the timescales for heat conduction were too long to explain the rapid production of a large volume of magma by melting of hydrous lithospheric mantle and proposed that the

Paraná basalts were generated by adiabatic decompression melting beneath a 45 km lithospheric lid. Such a model, however, does not entirely account for the wide range of trace-element signatures present in the majority of Paraná basalts and an alternative explanation is that these contain melts derived from both the convecting and lithospheric mantle [16]. Further east, the earliest plume-derived melts were generated in the Etendeka region of NW Namibia at 132.85 Ma [5], at very high pressures (~ 5 GPa [62]). Subsequent 132 Ma plume-derived melts formed at pressures as low as 1.5 GPa and are evidence that adiabatic decompression melting and lithospheric thinning had increased significantly on the eastern fringe of the plume impact zone [19]. Estimates of the amount of adiabatic decompression melting beneath Etendeka are quite high (12–22% [19]) but, somewhat surprisingly, the volume of melt emplaced in the region is relatively small. The apparent south-east migration of tholeiitic magmatism in the Paraná–Etendeka province proposed by Turner *et al.* [2] has been disputed by Renne *et al.* [5] who suggested that volcanic activity decreased in age northwards and reflected the south–north direction of rifting in this part of the South Atlantic. This northward decrease in age of volcanism along the South American coastal margin, also correlates with the internal configuration of magma types within the lava pile [63]. Our new $^{40}Ar/^{39}Ar$ age determinations for alkaline igneous rocks from eastern Paraguay (Table 1) support the proposition that, in general, magmatism migrated south-eastwards across the province. This may relate to the north-westerly drift of the lithosphere over the plume axis; reconstructions of plate motions suggest that the centre of the present-day Tristan plume was located beneath the eastern margin of the Paraná sedimentary basin, near the Ponta Grossa ‘Arch’, at 130 Ma and that the lithosphere was moving at ~ 3.5 cm yr^{-1} in a north-westerly direction [61]. The amount of Early-Cretaceous extension beneath the Ponta Grossa Arch is low ($\beta = < 1.5$ [64]), such that high amounts of adiabatic decompressional melting in the upwelling plume could only occur if the original thickness of the MBL was < 100 km.

(iv) *Crustal melting, 12 to 17 Myr after plume impact.* Some of the Paraná rhyolites (Chapeco and Aigüa Series, Fig. 2) are believed to represent anatectic melts of mafic lower crust, e.g. [65,66]. We note that these occur in regions where seismic velocities are low at depths > 100 km. This association is consistent with our conductive heating models which suggest that, in order to raise the temperature at the base of the crust, from the steady-state temperature of 700 °C to the melting temperature of the rhyolites (850 °C) in 15 Myr, the MBL would have to < 70 km thick. Nevertheless, the correlation of individual silicic units in Namibia and southern Brazil requires that their emplacement prior to continental break-up, unless unreasonably large magma volumes are inferred [67].

(v) *Extension and heating of metasomatised lithosphere 11 to 17 Myr after plume impact*. The alkaline complexes along the South American and African coastal margins were generated at the same time as the main pulse of tholeiitic volcanism, but just before the emplacement of the major dyke swarms (Fig. 6). It seems that a small amount of extension, in addition to conductive heating, was required to form these. A similar mechanism may have been responsible for the final phase (127.5 Ma) of alkaline igneous activity in eastern Paraguay, which was remote (475 km) from the continental margin, and in a different region from that affected by pre-flood-basalt alkaline magmatism at 145 Ma. The late-stage melting of metasomatised cratonic lithosphere appears to have been related to the development of the Sapucai–Villarica rift which may in turn have been caused by extensional forces associated with the final opening of the South Atlantic. Thermal weakening (due to conductive heating) of the lower crust and upper lithospheric mantle of the Rio de la Plata craton may also have promoted this extension. The 127.5 Ma small-fraction, volatile-rich melts occur in the same region as early tholeiitic basalts (Fig. 6) and we regard their presence as evidence against wholesale melting of ‘wet’ lithospheric mantle during CFB genesis; if this had occurred beneath the western margin of the Paraná basin at ~ 139 Ma [15] then it is unlikely that there would be readily-fusible, enriched veins present in the SCLM at 127.5 Ma.

6. Conclusions

(i) High-precision $^{40}Ar/^{39}Ar$ age determinations of volatile-rich alkaline igneous rocks in eastern Paraguay indicate that small-fraction melt generation occurred in two discrete episodes at 145 and 127.5 Ma. The earliest phase of this alkaline igneous activity pre-dates that from elsewhere in the Paraná–Etendeka large igneous province by at least 11 Ma.

The youngest phase (127.5 Ma) is similar in age to the oldest sea floor at these latitudes [49].

(ii) In the Paraná–Etendeka province, the early-phase of both alkaline and tholeiitic igneous activity occurred in the west. Alkaline igneous activity predates tholeiitic volcanism by 6 Myr. The sequence and timescale of igneous activity in this large igneous province is consistent with the findings of laboratory experiments and numerical models undertaken to simulate the impact of mantle plume starting heads.

(iii) Radiogenic-isotope ratios (εNd = −11 to −22) and trace-element concentrations (e.g. K_2O and Ni are up to 8.5 wt.% and 325 ppm, respectively) suggest that the source of the Paraguayan alkaline melts was phlogopite-bearing peridotite in the lithospheric mantle.

(iv) The early-phase Paraguayan alkaline igneous complexes are located on the margins of thick lithosphere. Simple conductive heating models suggest that, following mantle plume impact, a 10 km thick metasomatised zone at the base of the lithosphere would melt within just a few million years. The late phase of alkaline activity in Paraguay occurs in a rift zone away from the margin of the Rio de la Plata craton. Lithospheric extension, aided by thermal weakening, may have been important in triggering this small-fraction melting event and also those beneath other parts of the Paraná–Etendeka LIP (e.g. Damaraland, Brazilian and Uruguayan coastal margins).

(v) The total duration of igneous activity in the Paraná–Etendeka province appears to have been much greater than that in other Phanerozoic LIPs, such as the Deccan and Siberia. There are, however, significantly more high-precision ages published for the Paraná–Etendeka province (Fig. 1) and the long duration of igneous activity may be an artefact of the available ages rather than a consequence of different melt-generation processes associated with impacting mantle-plume starting-heads.

(vi) Despite the fact that volatile-rich, alkaline-igneous activity has been observed in almost all Phanerozoic LIPs, only a few high-precision ages have been published. These suggest that, with the exception of the Paraná–Etendeka LIP, small-fraction melt generation both pre- and post-dates the peak of tholeiitic volcanism by up to 3 Myr [7,8,68–70]. The large time interval (∼ 10 Myr) that separates early alkaline igneous activity from the peak of tholeiitic volcanism in the Paraná–Etendeka LIP may reflect the widespread presence of thick lithosphere at the time of plume impact. The absence of 'old' ages for alkaline magmas from LIPs may be due to the susceptibility of early-phase small-fraction volatile-rich melts to burial by thick successions of tholeiitic basalts and it may be extremely fortunate that we have been able to sample these in the Paraná–Etendeka LIP.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.epsl.2006.08.004.

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