# The alkaline-carbonatite complex of Jacupiranga (Brazil): magma genesis and mode of emplacement

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#### Abstract

A comprehensive study including new field, petrological and geochemical data is reported on the Jacupiranga alkaline-carbonatite complex (133-131 Ma) which, together with other alkaline complexes, occurs in southern Brazil and is coeval with- the Paraná CFB province. It consists of a shallow intrusion (ca. 65 km<sup>2</sup>) in the Precambrian crystalline basement, and can be subdivided in two main diachronous plutonic bodies: an older dunite-gabbro-syenite in the NW and a younger clinopyroxenite – ijolite (s.l.) in the SE, later injected by a carbonatitic core (ca. 1 km<sup>2</sup>). An integrated petrogenetic model, based on bulk rock major and trace element analyses, mineral chemistry and Sr-Nd-Pb-C isotopic data, suggests that the two silicate intrusions generated from different mantle-derived magmas that evolved at shallow level (2-3 km depth) in two zoned cupshaped plutonic bodies growing incrementally from independent feeding systems. The first intrusion was generated by OIB-like alkaline to mildly alkaline parental basalts that initially led to the formation of a dunitic adcumulate core, discontinuously surrounded by gabbroic cumulates, in turn injected by subanular syenite intrusive and phonolite dykes. Nephelinitic (+ melilite) melts – likely generated deep in the lithosphere at ≥ 3 GPa- were the parental magmas of the second intrusion and gave rise to large coarse-grained clinopyroxenite ad- to meso-cumulates, in turn surrounded, and partially cut, by semi-annular fine-layered melteigite-ijolite-urtite ortho-cumulates. The available isotopic data do not evidence genetic links between carbonatites and the associated silicate intrusions, thus favouring an independent source from the mantle. Moreover, it may be suggested that, unlike gabbro-syenites and carbonatites, mostly generated from lithospheric mantle sources, the parental magmas of the ijolite-clinopyroxenite intrusion also record the influence of sublithospheric (plume-related?) geochemical components.

#### 1. Introduction

Among the nearly 500 alkaline-carbonatite igneous rock associations of the world (Woolley and Kjarsgaard, 2008), the Jacupiranga Complex in southern Brazil is quite famous having been the object of several investigations, most of which focused either on the general aspects of the complex or on its carbonatite core which represents less than 1% in volume with respect to the outcropping silicate intrusives (Melcher, 1966; Gaspar and Wyllie, 1982; 1983a; 1983b; 1987; Roden et al., 1985; Morbidelli et al., 1986; 1995; Germann et al., 1987; Huang et al., 1995; Costanzo et al., 2006; Gomes et al., 2011; Salvioli-Mariani et al., 2012). According to field and petrographic observations, this complex is formed by different intrusions and fundamental questions concerning the petrogenesis and the mutual relationships between the different lithological units need to be addressed. This involves also the origin of carbonatite rocks, which have been considered in turn generated either by direct partial melting of carbonated mantle sources (Gittins, 1989; Yaxley et al., 1991; Huang et al., 1995), liquid immiscibility (Andersen, 1988; Le Bas, 1989; Beccaluva et al., 1992; Gomes et al., 2011) or extreme fractional crystallization processes (Wyllie, 1987; Comin-Chiaramonti et al., 2014). Some of these questions can be properly addressed only when the studied magmatic complex can be considered -at least in part- a closed petrological system, i.e. containing all its lithological components. In this regard, the Jacupiranga Complex represents a suitable casestudy for the presence of rocks related to distinct magma pulses which include mafic compositions approaching those of primary melts, felsic compositions and cumulate rocks resulting from differentiation processes. Moreover, the nearly coeval age of Jacupiranga (and other neighbouring alkaline occurrences) with Paraná Continental Flood Basalts (CFB), makes this magmatism of particular interest for the tectonomagmatic evolution of this sector of Western Gondwana, providing complementary information on the sources of alkaline magmas with respect to those of CFB (cf. Comin- Chiaramonti and Gomes, 2005 and Comin-Chiaramonti et al., 2011). Similar co-existence of alkaline complexes with tholeitic lavas has been observed in CFB provinces such as Karoo (Ellam and Cox, 1991; Jourdan et al., 2007), and Deccan (Sen et al., 2009), indicating that extremely different magmas may be nearly coeval at the same site. In this regard, plume-lithosphere interactions have been called upon for the genetic relationships between Paraguay alkaline complexes and the Paraná magmatism (Gibson et al., 2006) and, more in general, attention has been increasingly drawn on the alkaline magmatism and mantle plume activity in both oceanic and continental environments (Safonova and Santosh, 2014; Pirajno, 2015).

In this paper we present a complete set of georeferenced petrographical, geochemical and mineralogical data on the multiple intrusions of the Jacupiranga Complex, including new bulk rock major and trace element analyses (XRF, ICP-MS), mineral chemistry data (EMPA) and Sr-Nd-Pb-C isotopic data (TIMS, EA-IRMS). Our results provide a complete and detailed picture of the whole Jacupiranga magmatic system for the first time, and are aimed 1) to investigate the genetic relationships between the different rock types; 2) to clarify the petrogenesis of carbonatites; 3) to highlight the significance of this complex in the regional tectono-magmatic framework.

# 2. Geological setting

The Jacupiranga Complex crops out at the contact between granodiorites and micaschists of the Precambrian basement and, like other alkaline-complexes, lies along the Guapiara fault system, at the northern flank of the Ponta Grossa Arch (Beccaluva et al., 1992; Morbidelli et al., 1995; Comin-Chiaramonti and Gomes, 2005; Ruberti et al., 2005; Gomes et al., 2011; Azzone et al.,

2013). This uplifted structure has been affected since Early Cretaceous by northwest/southeast tensional fractures which have controlled the emplacement of alkaline magmas as well as dyke swarms related to the Paraná flood basalts (Piccirillo et al., 1989; 1990). The total thickness of the sedimentary/CFB cover on the Precambrian basement, together with the erosion rate estimate in the region, suggest an original depth of at least 1.5 km for the Jacupiranga intrusion (Melcher, 1966).

Several K/Ar and Rb/Sr datings indicate a maximum age span of 140-129 Ma (Amaral, 1978; Roden et al., 1985; Sonoki and Garda, 1988). This makes the Jacupiranga intrusion one of the oldest Phanerozoic alkaline-carbonatite occurrences of southern Brasil, nearly coeval with most Paraná-Etendeka CFB that were emplaced in the chronological range 135-131 Ma (Stewart et al., 1996; Renne et al., 1996a; 1996b; Thiede and Vasconcelos, 2010; Janasi et al., 2011). Recent thermochronological investigations, based on apatite fission tracks, suggest post-magmatic cooling and denudation of the Jacupiranga complex since 90 Ma (Soares et al., 2015). Rifting that dismembered the CFB/sedimentary cover, continued in several pulses accompanying the continental break-up and the south Atlantic opening (Chang et al., 1992; Strugale et al., 2007).

The Jacupiranga complex covers an area of ca. 65 km² elongated in NW-SE direction and may be subdivided in two main distinct intrusions which are partially interfingered at the contact, also in relation to brecciation processes. The older intrusion, in the the NW half part of the complex, consists of an early dunite core (widely serpentinized owing to in situ supergene processes) surrounded by gabbro-syenite lithologies. The mafic rocks consist of alkali-gabbros and syeno-diorites representing early solidified magmas outcropping discontinuously mainly at the intrusion border, whereas fine- to coarse-grained syenites represent late differentiated magmas injected peripherically respect to dunites. Alkali-basaltic and phonolitic dykes are found in the northernmost part of this intrusion or in the neighbouring Precambrian basement. The second intrusion, in the SE half part, mostly consists of a large massive body of clinopyroxenite (jacupirangite) which is in turn cut by a half ring of fine-layered urtite-ijolite-melteigite; the latter more mafic lithotype is concentrated, as a semiannular shell, around the clinopyroxenite body. Both clinopyroxenites and

ijolites (*s.l.*), as well as some ankaratritic dykes, cut the first intrusion, providing relative chronological evidence of the two magmatic events. The final magmatic episode of the complex is represented by carbonatite, which outcrops as an elongated core (less than 1 km across) at the centre of the clinopyroxenite body; it is formed by variably textured sövite (calcite-carbonatite) and minor rauhaugite (dolomite-carbonatite) rocks representing a nearly vertical (70-90°) composite plug resulting from at least six discrete units, which become younger northward (Gaspar and Wyllie, 1983a; 1983b; Alves, 2008; Barbosa Landim et al., 2010). Several carbonatite veins cross-cut the surrounding clinopyroxenite rocks, that sometimes are found as xenoliths included in the carbonatite body. A reaction facies has been observed between carbonatites and the surrounding clinopyroxenite, locally reaching the width of two metres; it consists of a millimetric to centimetric alternation of carbonate and silicate-rich bands, with the latter mainly composed of brown mica, amphibole and oxides (Gaspar, 1992). As synthetized in the geological cross-sections of Fig. 1, lithological contacts appear to be mostly subvertical as indicated by dyke geometry and geophysical data that evidence high angle boundaries traceable down to several hundred meters in depth (Alva-Valdivia and López-Loera 2011; Shukowsky et al., 2003).

The chronological relationships resulting from field observations seem to be supported by K/Ar ages on brown micas (Sonoki and Garda, 1988), which cluster around  $133 \pm 3$  Ma and  $130 \pm 3$  Ma for the first and second silicate intrusion, respectively. Moreover the Rb/Sr isochron for carbonatites gives an age of  $131 \pm 3$  Ma (Roden et al., 1985). The timing of the different events has been recently confirmed by more accurate Ar-Ar radiometric datings, which yield an age of  $134.9 \pm 0.7$  Ma for a syenite of the oldest intrusion and 131 for both clinopyroxenite and carbonatite (Chmyz et al., 2015). A further time limit is provided by in situ U-Pb datings on zirconolite in Jacupiranga carbonatites that yield an age of  $125 \pm 6$  Ma (Wu et al., 2010).

#### 3. Materials and methods

The least altered and most representative magmatic lithotypes were selected out of 150 specimens and powdered in an agate mill. Major and trace elements (Ni, Co, Cr, V, Rb, Sr, Ba and Zr) were analysed by X-ray fluorescence (XRF) on powder pellets, using a wavelength-dispersive automated ARL Advant'X spectrometer at the Department of Earth Sciences, Ferrara University. Accuracy and precision for major elements are estimated as better than 3% for Si, Ti, Fe, Ca and K, and 7% for Mg, Al, Mn, Na; for trace elements (above 10 ppm) they are better than 10%. REE, Y, Hf, Nb, Ta, Th, and U were analysed by inductively coupled mass spectrometry (ICP-MS) at the Department of Earth Sciences, Ferrara University, using a Thermo-Scientific X-Series. Accuracy and precision, based on the replicated analyses of samples and standards, are estimated as better than 10% for all elements, well above the detection limit.

Mineral compositions were obtained at the University of Modena using a ARL-SEMQ electron microprobe (fitted with three wavelength dispersive spectrometers) at an accelerating voltage of 15 kV, and specimen current of 20 nA, and at Centro Interdipartimentale Strumentazioni per Analisi Geomineralogiche (CISAG), University of Naples, using an Oxford Instruments Microanalysis Unit equipped with an INCA X-act detector and a JEOL JSM-5310 microscope at an accelerating voltage of 15 kV 50–100 mA filament current. Natural silicates and oxides were used as standards in both laboratories.

Sr-Nd-Pb isotopic analyses on bulk rock was carried out at the CNR-IGG of Pisa, where Sr-Nd-Pb isotopic compositions were determined by Thermal Ionization Mass Spectrometry (TIMS) using a Finnigan MAT 262 multi-collector mass-spectrometer following the methods described in Saccani et al. (2013) and Natali et al. (2016). In this laboratory replicate analyses of the NBS 987 standard gave an average <sup>86</sup>Sr/<sup>88</sup>Sr value of 0.710253±13 (2σ, n=30), whereas the isotopic standard JNdi-1 (Tanaka et al., 2000) gave an average <sup>143</sup>Nd/<sup>144</sup>Nd value of 0.512098±8 (2σ, n=25). As concern Pb, replicate analyses are accurate to within 0.025% (2σ) per mass unit, after applying mass discrimination correction of 0.15±0.01 per mass unit relative to the NBS 981 reference composition of Todt et al. (1993).

C elemental and isotopic analyses have been carried out on bulk rock powders at the University of Ferrara using a Vario Micro Cube Elemental Analyzer (EA) coupled with an Isoprime 100 Isotope Ratio Mass Spectrometer (IRMS). The elemental precision estimated by repeated standard analyses and accuracy estimated by the comparison between reference and measured values were in the order of 5 % of the absolute measured value. Uncertainties increase for contents approaching the detection limit (0.001 wt%). Carbon isotope ratios are expressed in the standard ( $\delta$ ) notation in per mil (%) relative to the international Vienna Pee Dee Belemnite (V-PDB) isotope standard. The  $\delta^{13}$ C values were characterized by an average standard deviation of  $\pm 0.1\%$  defined by repeated analyses of the abovementioned standards (see Natali and Bianchini, 2015 for additional procedural details).

# 4. Petrography and classification

Major and trace element analyses on selected Jacupiranga rocks are reported in Table 1. Chemical classification for silicate lithotypes has been performed using the R1-R2 diagram of De La Roche et al (1980; Fig. 2). The nomenclature of Irvine (1982) has been used for cumulitic textures. In the R1-R2 classification diagram (Fig. 2) rocks of the two main intrusions show distinct differentiation trends: the first corresponds to an alkaline suite, the second to an extremely alkaline silica-undersaturated suite. Table 2 reports the main mineral assemblages of Jacupiranga rocks.

Modal composition (wt%) of the main rock groups estimated by mass calculation between bulk rock and constituent mineral analyses is reported in Table 3 and Fig.3. Relevant indexes (see Frost and Frost, 2008) such as the AI (alkalinity index = molar Al – [Na+K]) and the FSSI (feldspathoid silica saturation index = normative Q-[Lc+2\*{Ne+Kp}]/100) are also reported in Fig. 3 to emphasize serial differences between rocks of the two silicate intrusions. The petrographic description of the Jacupiranga rocks is reported below and illustrated in microphotographs of Figs. 4 and 5.

The north-western intrusion includes alkali gabbros, syeno-diorites and syenites around dunites. Dunites (Mg# 0.88-0.84) are mostly serpentinized; the freshest samples show adcumulitic texture and consist of olivine (> 95%), chromite, and scarce intercumulus brown mica and clinopyroxene. Alkali-gabbros (Mg# 0.59-0.56; normative nepheline 0.4-2.0 wt%) are characterized by hypidiomorphic medium-grained texture and are made up of euhedral olivine (<10 wt%), abundant clinopyroxene (25-29 wt%) and plagioclase (31-40 wt%) followed by magnetite + ilmenite (< 10 wt%), brown mica (13-19 wt%) and alkali-feldspar (6-13 wt%); apatite (1-2 wt%) is a widespread accessory phase. Syeno-diorites (Mg# 0.51; normative nepheline ca. 6 wt%) display hypidiomorphic texture consisting of early-crystallized euhedral plagioclase and clinopyroxene within an interstitial, sometimes fluidal, assemblage made of brown mica, alkali feldspars, opaques and accessory apatite. Syenites (Mg# from 0.48 to 0.30) are characterized by variable degree of silica saturation; most samples display normative nepheline (up to 12 wt%), whereas a few quartz bearing samples outcrop exclusively at the northernmost border of the complex. Syenites are characterized by inequigranular coarse- to fine-grained texture with dominant large (sometimes centimetric) crystals of alkali feldspar (36-74 wt%), minor euhedral plagioclase (0-46 wt%), clinopyroxene (5-16 wt%), opaques (1-6 wt%), brown mica (0-10 wt%) and amphibole (0-14 wt%). Nepheline, nosean or quartz are interstitial phases in the least (e.g. JC107, JC112, nepheline up to 15 wt%) and most (e.g. JC119, JC120, quartz up to 4 wt%) silica-saturated rocks, respectively. Apatite and titanite are common accessory phases. Phonolitic dykes represent the most evolved magmatic compositions (Mg# 0.18-0.14) of the first intrusion, showing invariably high silicaundersaturation (normative nepheline 22-28%) and attaining a peralkaline character ([Na+K]/Al mol up to 1.04). They are characterized by phenocrysts of alkali feldspar, clinopyroxene, amphibole and minor plagioclase, followed by magnetite, apatite and titanite within a holocrystalline groundmass made of the same phases plus nepheline, aegirin-augite clinopyroxene, sodalite, and melanitic garnet.

The south-eastern intrusion includes clinopyroxenites and rocks of the ijolite-melteigite-urtite group. This suite shows a higher alkalinity and silica-undersaturation which is modally reflected in the systematic presence of nepheline (± melilite), and in the absence of plagioclase. Clinopyroxenites (Mg# 0.75-0.53), known as "jacupirangites", have generally massive (occasionally weakly layered) ad- to meso-cumulitic inequigranular texture with cumulus clinopyroxene (up to 5 mm across) and rare olivine; inter-cumulus phases (from 5 to 20 wt%) are mainly represented by magnetite, brown mica, apatite, with perovskite and ilmenite as accessory phases. No carbonatitic ocelli were observed in this lithology, whereas rare amphibole is found close to the carbonatite intrusion. Ijolites, melteigites and urtites are distinguished by their nepheline/clinopyroxene ratio and are characterized by ortho-cumulitic layered textures. Melteigites constitute a semiannular shell around the clinopyroxenite body and are finer-grained with respect to ijolites and urtites which are often characterized by medium- to coarse-grained textures. The main cumulus phase is represented by clinopyroxene (up to 75 wt% in melteigites), rarely including sporadic olivine, whereas the intercumulus phases consist of magnetite (1-9 wt%), brown mica, nepheline (up to 62 wt% in urtites) and alkali feldspar (up to 27 wt% in urtites). Apatite, perovskite, occasionally titanite, and exsolved kalsilite are distinctive accessory minerals of all these rocks. Celsian is observed in urtites. Some ijolite, melteigite, and urtites are characterized by significant presence of melilite (0-18 wt% in melteigites, 0-3 in ijolites wt%, 0-7 wt% in urtites). Of note, four melteigites having melilite above 10 wt% could be classified as melilitolites (Dunworth and Bell, 1998). The ankaratrite dyke JC 111D is slightly porphyritic with clinopyroxene phenocrysts set in a microgranular holocrystalline groundmass made of clinopyroxene, brown mica, magnetite, interstitial feldspathoids of the noseana-sodalite group and alkali feldspar together with titanite and apatite as accessory phases.

Carbonatites (mostly sövites), intruding the clinopyroxenite body, show variable textural and mineralogical features (Mitchell, 1978; Gaspar and Wyllie, 1983a; 1983b; 1987; Brod et al., 2001; Alves, 2008). They are coarse- to medium-grained (grain size between 0.3 and 7 mm) and

sometimes banded. The main mineral assemblages of Jacupiranga carbonatites include calcite, dolomite, apatite and magnetite; olivine, phlogopite, magnesian ilmenite, titanian clinohumite, pyrochlore, Pb-carbonates, barite sulfides, baddeleyite, zircolonite and calzirtite are accessory phases. The complex aureola around the carbonatite intrusions has been described in detail by Morbidelli et al. (1986) and Gaspar (1992).

#### 5. Mineral chemistry

Mineral chemistry data of the two main silicate intrusions of Jacupiranga are discussed in the following section; the extensive data set is reported in the supplementary section of the journal repository including Supplementary Table 1 and Supplementary Figs. 2-5.

#### 5.1 Dunite-gabbro-syenite intrusion

Olivine is present in the ultramafic to mafic rocks of this intrusion with Fo 82.6-87.7 in dunites and Fo 46.7-57.0 in gabbros. NiO is positively correlated with Fo, and decreases from 0.82 wt% in some dunites to < 0.1wt% in gabbros. Spinel of dunites is represented by chromian titanian-magnetite, with a chromite molar component of 26.3-43.7 mol%. In gabbros and syeno-diorites, spinel is represented by magnetite with ulvospinel up to 43.6 mol% and chromite up to 1.2 mol%. Ilmenite is usually associated with magnetite in nearly all lithologies. Clinopyroxene varies from Cr-bearing (Cr<sub>2</sub>O<sub>3</sub> up to 1.2 wt%) diopside in dunites to augite in gabbros up to aegirine-augite (Na<sub>2</sub>O up to 4.3 wt%; FeOtot up to 20 wt%) in some syenitic and phonolitic rocks (Supplementary Fig. 2). Feldspars are represented by plagioclase and alkali feldspar. Plagioclase composition is An<sub>46-20</sub> in gabbros and An<sub>41-16</sub> in syeno-diorites. Alkali-feldspar is in the range Ab<sub>39-21</sub> Or<sub>52-75</sub> in gabbros, Ab<sub>44-35</sub> Or<sub>50-60</sub> in syeno-diorites, Ab<sub>59-31</sub> Or<sub>39-68</sub> in syenites; perthitic exsolution is observed in syenites (Or<sub>87</sub> Ab<sub>13</sub> and Ab<sub>95</sub> Or<sub>2</sub>) and in phenocrysts of phonolites (Or<sub>94-88</sub> Ab<sub>6-12</sub>; Ab<sub>99-97</sub> Or<sub>0-1</sub>)

(Supplementary Fig. 3). Feldspathoids consist of nepheline in peralkaline phonolites and in the most silica-undersaturated syenites, where it is associated with minor haüyne (SO<sub>3</sub> 4-7 wt%) and sodalite (Cl 7.6 wt%). Nepheline (Ne<sub>74-79</sub>Ks<sub>19-17</sub>) is generally characterized by Si excess (Q<sub>3-6</sub>) respect to the idealized composition, with calcium decreasing from syenites (CaO 0.50-0.45 wt%) to phonolites (CaO < 0.2 wt%). Brown mica is ubiquitous in all the lithologies of this intrusion, including the dunite cumulates. Its composition varies from nearly pure phlogopite in dunites (Mg/Fe up to 12), to titan-phlogopites containing a significant annite component (Mg/Fe between 2.0-1.1; TiO<sub>2</sub> up to 7.6 wt%) in gabbros, syeno-diorites and syenites, to biotite in phonolites (Mg/Fe down to 0.3) (Supplementary Fig. 4). Amphibole consists of calcic amphibole (Na+K>0.5 and Ti<0.5 a.p.f.u.) and can be classified as edenite in dunites, edenite to magnesiohastingsite in syenites and hastingsite in phonolites on the basis of the Leake et al. (1997) scheme (Supplementary Fig. 5); this compositional variation, characterized by decreasing Mg#, is coherent with that of coexisting clinopyroxene, as shown in Supplementary Fig. 2. Accessory minerals are represented mainly by apatite showing fluor- and REE-rich compositions in the felsic rocks, where it is in association with titanite and sporadic melanitic garnet. Other rarer accessory minerals analyzed in syenites and peralkaline phonolites are Fe-Cu sulfides, zircon, baddeleyite, perrierite, thorurite, thorite, barite, wöhlerite/rosenbuschite, fluorite and minerals of huttonite/monazite series.

#### 5.2 Clinopyroxenite - ijolite (s.l.) intrusion

The rocks of this intrusion mainly consist of clinopyroxene, brown mica, magnetite, nepheline and perovskite. It has to be emphasized, the presence of melilite (in addition to nepheline) in the most silica-undersaturated ijolite/melteigite/urtite rocks. *Clinopyroxene* shows a remarkably homogeneous diopside composition, with comparatively higher (respect to the rocks of the first intrusion) TiO<sub>2</sub> (up to 3.3 wt%) and Al<sub>2</sub>O<sub>3</sub> (up to 6.6 wt%) and lower Na<sub>2</sub>O (down to 0.2 wt%); Supplementary Table 1 and supplementary Fig. 2). It is worth noting that in clinopyroxenites,

cumulus clinopyroxene with Mg# 83.2-76.8 coexists with sporadic olivine with Mg# 82.6-79.6, and rare Mg-hastingsite amphibole with Mg# 80.0-82.0 (Supplementary Fig. 5). Melilite, characterized by solid solution of Akermanite<sub>66.9-60.1</sub>, soda-Melilite<sub>31.4-37.3</sub> and soda-Gehlenite<sub>0.6-2.8</sub>, is a relatively early crystallized phase of melteigites, ijolites and urtites belonging to the most silica undersaturated suite. The Mg# ranges from 67.6 to 83.6 and has a composition similar to that of Lages melilites (Traversa et al., 1996) and other melilite-bearing nephelinites worldwide (Melluso et al., 2011a; Wiedenmann et al., 2010). Feldspathoids consist of nepheline and kalsilite. The nepheline compositions have Ne<sub>70-77</sub>Ks<sub>29-20</sub>, and are sometimes accompanied by nearly pure kalsilite. The CaO decrease from 1.4 wt% in melteigites to 0.1 wt% in urtites, whereas the silica excess  $(Q_{<4})$ , is generally low, differently from the nephelines of the first intrusion, and typical of nepheline of highly silica undersaturated rocks (Wilkinson and Hensel, 1994). Brown mica is widespread in all lithologies of the suite, and is generally a barium- and titanium-rich phlogopite (BaO up to 10 wt%, TiO<sub>2</sub> up to 8.6 wt %). With respect to the brown mica of the first intrusion, these phlogopites are comparatively richer in Al<sub>2</sub>O<sub>3</sub> (in addition to Ba) and poorer in SiO<sub>2</sub>, in relation to the extreme silica-undersaturation of the system and the consequent coupled substitution Ba+Al<sup>IV</sup> = K+Si. In the urtite sample JC79 brown mica displays a peculiar composition characterized by an extreme enrichment of iron components (Supplementary Fig. 4). Magnetite is by far the predominant Fe-Ti oxide; in clinopyroxenites, melteigites and ijolites the ulvospinel component is in the range 20-48 mol%, whereas in urtites ulvospinel component is down to 11 mol%. Perovskite is ubiquitous throughout this suite and typically contains significant amount of REE. Celsian and stronalsite are observed in the urtites JC87, JC89 and JC91, similarly to that observed in melteigites at Turiy (cf. Dunworth and Bell, 2003). Celsian is a featuring phase also in other melilite nephelinites elsewhere (cf. Melluso et al., 2011a; 2011b) Accessory minerals are represented mainly by fluorine-rich apatite and Fe-Cu-Pb sulphides.

## 5.3 Comparison with other alkaline-carbonatite complexes

Taking into consideration the existing literature which relates carbonatites and the coexisting silicate rocks provided by Wolley and Kjarsgaard (2008) and the relative categorization, the rock association observed at Jacupiranga including both peridotites and pyroxenites among ultramafic rocks, ijolite (s.l.) grading to melilitolites, syenites and carbonatites appear to be quite rare. Best analogues are represented by the alkaline-carbonatite occurrences of Kovdor (Kola Peninsula, NW Russia; Verhulst et al., 2000), Gardiner (East Greenland; Nielsen, 1980) and Sung Valley (NE India; Srivastava and Sinha, 2004) that are ring complexes showing comparable size and similar parageneses. In particular, the presence of melilite in the rocks of the second intrusion deserves to be noted, because according to the statistical observation of Wolley (2003), Wolley and Church (2005) and Wolley and Kjarsgaard (2008) this mineral is common in silicate rocks associated to extrusive carbonatites, but rare in the plutonic analogues where wollastonite is more frequent.

## 6. Parental magmas and fractionation processes

The two intrusions show remarkably different silica undersaturation, which is reflected in distinctive modal and normative mineral assemblages: the gabbro-syenite suite of the first intrusion contains ubiquitous modal plagioclase and moderate normative nepheline, whereas the clinopyroxenite-ijolite (s.l.) suite does not contain modal plagioclase and is characterized by abundant normative and modal nepheline as well as modal melilite. These critical differences are clearly illustrated by the diopside-nepheline-akermanite-silica systems (Onuma and Yamamoto, 1976) where the normative compositions of the two intrusive suites plot on one or the other side of the diopside-nepheline join (Fig. 6), as well as by systematic differences in the mineral chemistry of constituent phases described above. Coherently, compositional differences also characterize the relative ultramafic cumulates, which are dunites in the first intrusion and clinopyroxenites in the second. The identification of parental magmas in plutonic complexes is usually a difficult task

owing to the extensive cumulitic processes that generally obliterate the liquid line of descent represented by parent and differentiated magmas during fractional crystallization processes. For the Jacupiranga intrusions, mafic dykes and fine-grained border facies, may help to constrain parental magma compositions. Moreover, the presence of both ultramafic cumulates and felsic rocks in the two main intrusions allow to assume that the final differentiation processes approached closed system conditions, and therefore reconstruction of the various magmatic stages can be tested by petrological and geochemical modelling. Such modelling has been tested using: 1) least square mass balance calculations involving the major element composition of bulk rock and constituent minerals; 2) phase equilibria estimates by the PELE software (Boudreau, 1999; Natali et al., 2011); 3) incompatible element best fit between PELE results and observed differentiate compositions. Model parameters for PELE were oxidized QFM +1/+2 and low pressure (0.1-0.3 GPa) conditions, since shallow fractionation is suggested - beside geological considerations - by barometric estimates on clinopyroxene-rock pair (Putirka et al., 2008; Masotta et al., 2013) that constrain the pressure crystallization generally below 0.4 GPa. Moreover, the low pressure conditions seem to be coherent with the observed association of nepheline and melilite in the second intrusion, which is relatively common in alkaline volcanic/subvolcanic occurrences and rarer in deep plutonic analogues (Yagi and Onuma, 1978; Wilkinson and Stolz, 1983; Wolley, 2003; Wolley and Church, 2005; Rass, 2008; Stoppa et al., 2016).

Model results have been synthesized and compared in Supplementary Table 2 together with the surface (%) estimates of the various lithologies obtained from the geological map of Fig. 1.

## 6.1 Dunite-gabbro-syenite intrusion

The rocks that could best approximate the least differentiated magma compositions of the first intrusion are represented by gabbros (samples JC 104 and JC 122) and basaltic dykes (sample JC 126 D), varying in affinity from alkaline (normative nepheline up to ~ 3 wt%) to mildly alkaline

(normative hypersthene up to 8 wt%). Preliminarily, least square mass balance calculations between whole rocks and constituent mineral phases were carried out in order to test the possibility that syenitic compositions could derive from the above mentioned least differentiated basic rocks by common fractionation processes; results indicate that syenites could result manly by removal of 4-7 wt% olivine, 25-30 wt% clinopyroxene, 26-38 wt % plagioclase, 15-19 wt % biotite and 5-6 wt % Fe-Ti oxides. The assumed parental rocks however, can not be considered the real primary magmas, owing to the low Mg# (0.55-0.59), low Ni abundances (less than 90 ppm), as well as low forsteritic olivine (Fo 46.7-57.0). To reach Mg# > 0.68, compatible with a direct mantle derivation (Green & Falloon, 2005), it is necessary to add 14-16 wt% of Fo 86-88 olivine to the above basic compositions (first column of Supplementary Table 2). Noteworthy, the olivine required to be added is in reasonable agreement both for the observed composition (Fo 82.6-87.7) and the estimated volume of cumulitic dunites in field (ca. 18%, third column of Supplementary Table 2). This also implies early crystallization temperature of olivine in dunites around 1300 °C, according to the Ford et al., (1983) geothermometer. Therefore, dunites can be considered the cumulitic solid fraction, whose removal from the inferred primary melts led to the least differentiated basic rocks observed in this intrusion. Results from the thermodynamic modelling by the PELE software indicate that shallow fractional crystallization processes from the observed least differentiated basic compositions could generate several liquid lines of descent (LLD, Fig. 2) leading to syenitic differentiates which correspond to a residual liquid fraction in the range of 15-25 wt% (second column of Supplementary Table 2). In this framework, peralkaline phonolitic dykes may represent the extreme differentiated melts segregated from the nepheline-syenite crystal mush. All the modelled trends are invariably dominated by fractionation of 5-12 wt% olivine (since ca. 1200 °C), 15-20 wt% clinopyroxene, 20-30 wt % plagioclase, 5-10 wt% Fe-Ti oxides, up to 5 wt% biotite, 5-15 wt% alkali feldspar (at 800-850 °C), and minor amounts of apatite and titanite (Fig. 7).

It appears that the silica saturation degree of differentiates does not depend from the specific fractionation process but could be, in principle, controlled by the alkalinity of the parent, with

nepheline-syenites and quartz-syenites derived from alkaline (e.g. gabbro JC 122) and transitional (e.g. alkali basalt JC126) basic magmas, respectively (Fig. 2, cf. Natali et al., 2013). However, silica oversaturated syenites, cogenetic with silica undersaturated felsic rocks in other alkaline igneous complexes has been attributed to crustal contamination processes that could be able to drive critically undersaturated magmas across the feldspar join and produce oversaturated rocks (Foland et al., 1993 and Landoll et al., 1994; Riishuus et al., 2008). Similar interpretation was also suggested by Gaspar (1989) for the Jacupiranga complex and may find additional support by the recent discovery of orthopyroxene-bearing gabbroic rocks, reported by Azzone et al. (2012).

Chondrite (Ch)-normalized REE and Primordial Mantle (PM)-normalized incompatible element distributions show a general increase from mafic to felsic rocks that is compatible with the above modelled fractionation processes (Fig. 8). Phosphorous and titanium invariably display negative anomalies related to removal of apatite and Fe-Ti oxides, whereas other elements such as strontium and barium bear either negative or positive anomalies depending on subtraction or cumulus of feldspar during the differentiation. This fractionation scheme is quantitatively confirmed in Fig. 9, where the trace element distribution of the extreme differentiates (syenites and peralkaline phonolites) may represent ~ 20 wt% liquid fraction after removal of the same solid compositions computed by PELE major elements modelling (second column of Supplementary Table 2); the low concentration of heavy REE in peralkaline phonolites conforms to the presence of melanitic garnet as fractionating phase, that preferentially incorporates heavy-REE (Gaspar et al., 2008).

The normalized patterns of gabbros and basalts, confirm the alkaline affinity of the basaltic magmas of this intrusion ( $La_n/Yb_n = 22-44$ ), displaying incompatible element distributions typical of Ocean Island Basalts (OIB, Sun and McDonough, 1989; Weaver, 1991; Hofmann, 1997), broadly corresponding to moderate melting degrees of fertile lherzolite at > 1.5 GPa (Green and Fallon, 2005).

6.2 Clinopyroxenite (jacupirangite) - ijolite (s.l.) intrusion

Due to the widespread cumulus processes occurred in this intrusion, particular caution has to be exerted for the identification of parental magmas, that should rely both on compositional and textural constraints. Some of the fine-grained melteigites (JC 81-82) and the ankaratrite dyke (JC111-D) may be indicative of parental liquids of this intrusion. Starting from these compositions (Mg# 0.65-0.67) least square mass balance calculations between whole rocks and constituent mineral phases indicate that the most differentiated urtite compositions could result from removal of a mineral assemblage mainly formed by 4-10 wt% olivine, 40-70 wt% clinopyroxene, 10-15 wt% phlogopite, 0-17 wt% melilite, 0-1 wt% oxides (first column of Supplementary Table 2). Thermodynamic modelling has been also performed by PELE software (Boudreau, 1999) in order to test the phase equilibria during fractional crystallization: all fractionation models (Fig. 10) are invariably dominated by crystallization of 50-60 wt% clinopyroxene which appears early in the sequence (since ca. 1200 °C), whereas olivine is subordinate (< 5-15 wt%; second column of Supplementary Table 2). This is in agreement with the composition of the clinopyroxenite cumulates where olivine is observed only as minor phase. The subsequent crystallizing phases are invariably represented by magnetite, biotite, perovskite, apatite and nepheline as the last crystallizing phase (at 750-800 °C). Occasional titanite and alkali feldspar also appear late in the crystallization sequence. Noteworthy, PELE modelling indicates melilite crystallization only for the most silica undersaturated starting compositions (e.g. melteigite JC82), where modal melilite, in addition to nepheline, characterizes the related fractionation trend. The calculated liquid lines of descent (LLD) fit well the magmatic evolution from melteigites to ijolites and urtites, the latter representing the extreme differentiates of both melilite-bearing and melilite-free trends (Fig. 2). Therefore model results indicate that, in spite of clinopyroxene accumulation, also melteigites (in addition to ankaratrite) provide coherent indications on the nature and phase equilibria of parental magmas that could be equated to nephelinites.

Ch-normalized REE and PM-normalized incompatible element distributions of rocks pertaining to the second intrusion are shown in Fig. 11. It can be observed that ijolites *s.l.* are characterized by K and Rb spikes in addition to a generally higher contents of all incompatible elements with respect to clinopyroxenites. This implies that the latter mostly reflect cumulus of clinopyroxene, whereas ijolitic (*s.l.*) orthocumulates are variably influenced by differentiation processes: melteigites show various extent of clinopyroxene accumulation, ijolites and especially urtites display increasing abundance of residual liquids. Therefore, from the incompatible element perspective, the only plausible melt representative is the ankaratrite JC111-D that is characterized by remarkable LREE enrichment and La<sub>N</sub>/Yb<sub>N</sub> of 44, closely approaching the composition of nephelinitic rocks from other Brazilian alkaline occurrences (Traversa et al., 1996; Gomes et al., 2011; Bongiolo et al., 2015). A mafic dyke composition from the Jacupiranga complex closely resembling ankaratrite JC111-D (Fig. 7) has been recently reported by Menezes et al. (2015). The trace element modelling, reported in Fig. 12, indicates that the most differentiated urtites (e.g. JC74) may represent ~ 15 wt % liquid fraction after removal from ankaratrite JC111-D of the same solid compositions, computed by PELE major elements modelling (Supplementary Table 2).

On the whole, the incompatible element distribution of this intrusion conforms to that of strongly alkaline magmas (basanites, nephelinites and melilitites) commonly recorded in alkaline-carbonatite complexes worldwide (Bell, 2005; Woolley and Kjarsgaard, 2008). According to experimental results (Green & Fallon, 2005; Gudfinnsson and Presnall, 2005), these strongly silica-undersaturated magmas could be generated by very low (1-5 %) partial melting degree of fertile lherzolite (+ CHO) deep in the lithospheric mantle ( $P \ge 3$  GPa).

## 7. Isotope systematics

Isotopic analyses have been carried out on highly selected and representative samples from the various intrusions of the complex. Sr-Nd-Pb isotopic data are reported in Table 4 and plotted in Fig.

13 together with previous data on Jacupiranga from the literature (Huang et al., 1995). In the diagram <sup>87</sup>Sr/<sup>86</sup>Sr vs <sup>143</sup>Nd/<sup>144</sup>Nd of Fig. 13a it can be observed that the first and second intrusions have distinctive isotopic fingerprint. The gabbro-syenite association of the first intrusion shows 87Sr/86Sr between 0.7064 and 0.7093 and 143Nd/144Nd between 0.5123 and 0.5119, whereas clinopyroxenite-ijolite (s.l.) suite of the second intrusion displays <sup>87</sup>Sr/<sup>86</sup>Sr between 0.7047 and 0.7058 and <sup>143</sup>Nd/<sup>144</sup>Nd between 0.5126 and 0.5124. Lead isotopes also show distinct compositional trends for the two intrusions (Fig 13b): the first intrusion displays <sup>206</sup>Pb/<sup>204</sup>Pb between 17.3 and 17.9,  $^{207}Pb/^{204}Pb$  between 15.5 and 15.6 and  $^{208}Pb/^{204}Pb$  between 37.9 and 38.9, whereas the second intrusion shows <sup>206</sup>Pb/<sup>204</sup>Pb between 17.7 and 17.9, <sup>207</sup>Pb/<sup>204</sup>Pb between 15.4 and 15.5 and <sup>208</sup>Pb/<sup>204</sup>Pb between 38.0 and 38.4. Intra-suite compositional variation is relatively limited for the rocks of the second intrusion suggesting a clear comagmatic origin for the clinopyroxenite-ijolite association. On the other hand, significant compositional variations (beyond the notional mantle signatures) are observed for the gabbro-syenite association. The interpretation of this high isotopic variability is not straightforward but, in addition to source heterogeneity, it strongly suggests the significant occurrence of crustal contamination on magmas by the surrounding country rocks of the Precambrian basement (e.g. <sup>87</sup>Sr/<sup>86</sup>Sr = 0.709 in gabbro JC109 at the intrusion border), as also proposed for other Brazilian alkaline complexes (Azzone et al., 2016). Significant crustal contamination effects on magmas of the first intrusion could be also envisaged by the presence of cogenetic nepheline- and quartz-syenites, although further isotopic data are needed to confirm the diverse genetic hypotheses discussed in the previous sections. Carbonatites in turn have distinctly different isotopic signatures showing <sup>87</sup>Sr/<sup>86</sup>Sr between 0.7049 and 0.7054, <sup>143</sup>Nd/<sup>144</sup>Nd approaching 0.5126,  $^{206}\text{Pb}/^{204}\text{Pb}$  between 17.2 and 17.4,  $^{207}\text{Pb}/^{204}\text{Pb}$  approaching 15.4 and  $^{208}\text{Pb}/^{204}\text{Pb}$  between 37.7 and 37.9, indicating an independent origin with respect to the intruded clinopyroxenite-ijolite rocks, as already suggested by Huang et al. (1995).

On the whole, the Sr-Nd-Pb isotopic composition of Jacupiranga rocks is difficult to be addressed to a specific notional mantle end-members (Hofmann, 1997; Stracke, 2012), although it is

clearly displaced toward enriched mantle compositions (EM1 and EM2). Compared with the isotopic data reported by Hoernle et al. (2015) on the Tristan hot spot magmatism and the Paraná-Etendeka CFBs, the Jacupiranga gabbro-syenites and carbonatites show compositions plausibly derived from independent lithospheric sources, whereas the clinopyroxenite-ijolite intrusion displays a clear affinity with the "Gough component", considered characteristic of the initial plume activity (since 132 Ma) in western Gondwana.

In order to test the genetic hypothesis on carbonatites elemental and isotopic analysis of carbon has been carried out on Jacupiranga rocks and compared with those of the neighbouring and coeval alkaline-carbonatite complex of Juquiá for which carbonatite-silicate liquid immiscibility processes have been already established (Beccaluva et al., 1992). The results (Table 5 and Fig. 14) are significantly different for the two complexes. The Jacupiranga carbonatites (C = 11.7 wt%) shows a carbon isotopic signature ( $\delta^{13}$ C = -6.0 %) compatible with mantle values, whereas the associated gabbro-syenite and clinopyroxenite-ijolite (s.l.) intrusions have very low carbon content (usually lower than 0.1 wt%) and negative  $\delta^{13}$ C range (-13.5 to -24.6%). By contrast, the Juquiá complex show a gradual variation of both elemental and isotopic carbon composition from carbonatites ( $\delta^{13}$ C = -5.1 %, C = 6.9 wt.%) to the associated syenites and essexites ( $\delta^{13}$ C -7.6 to -13.5, C = 0.75-0.1 wt.%), up to the basanites ( $\delta^{13}$ C -18.2 ‰, C = 0.1 wt.%), in parallel with decreasing carbonatitic ocelli observed in silicate rocks. Therefore, for Jacupiranga the available data do not evidence genetic links between silicate rocks and carbonatite, suggesting an independent generation of carbonatites from the mantle. In Juquiá, on the contrary, the carbon systematics confirm that a gradual development of low pressure liquid immiscibility conditions (increasing ocelli) could eventually segregate carbonatite melts from the associated silicate magmas, as already proposed by Beccaluva et al. (1992). In this view, the extremely negative isotopic values recorded in silicate rocks of Jacupiranga silicate intrusions have to be considered the result of low-pressure degassing events in which the escaping CO<sub>2</sub> is <sup>13</sup>C-enriched (Pineau and Javoy, 1983; Des Marais and Moore,

1984). These processes that fractionate the carbon isotopes have been often observed in alkaline-carbonatite complexes (Nadeau et al., 1999).

## 8. A tentative model for the intrusion dynamics

Reconstruction of the magmatic intrusions at depth is not straightforward, since it is well known their highly variable mode of emplacement and geometry that depend on a number of factors such as composition, size and depth of plutonic bodies, nature of the host rocks, as well as tectonic regime. In particular, for many ring and Alaskan-type complexes (Chen et al., 2009; Oliveira et al., 2010; Andersson et al., 2016), the intrusion mechanisms can not be explained in terms of classic magma chambers where a homogeneous magma body undergoes fractional crystallization and crystal settling leading to mafic/ultramafic cumulates at the bottom and felsic differentiates at the top. More plausible models involve flow segregation (decoupling of crystals and melt; Menezes et al., 2015; Yamato et al., 2015; Natali et al., 2016) and crystal mush growing from the intrusion roof downward (Aarnes et al., 2008). Accordingly, an increasing number of papers have shown that many of shallow (<5 km depth) small-sized mafic intrusions, differently from major stratiform complexes, were emplaced in form of saucer- or cup-shaped plutonic bodies (Hansen and Cartwright, 2006; O'Driscoll et al., 2006; Polteau et al., 2008). Moreover, analogue experiments by Mathieu et al. (2008) and Galland et al. (2009; 2014) demonstrated that cup-shaped and cone-sheets intrusions could result by shear faulting at the tip of a nearly vertical feeding conduit under suitable stress regime and viscosity. In these complexes, the initiation of magma emplacement is plausibly characterized by multiple injections of dykes and sills, which progressively amalgamate at shallow level in the host rocks according to the "crack and seal model" (Bartley et al. 2008; Allibon et al., 2011). During such processes the combined effect of progressive crystallization and continuous magma inflation induce formation of mafic crystal mushes (mostly olivine and/or clinopyroxene) at

the centre of the plutonic body, whereas the remaining differentiated melts are forced to escape laterally through cone-sheets (Farahat and Helmy, 2006; Andersson et al., 2016).

Based on the above considerations, field data and petrogenetic modelling, we propose that the two Jacupiranga intrusions were emplaced according to the cartoons reported in Fig. 15. This model could satisfactorily account for the distinctive features of the Jacupiranga complex: 1) the subcircular concentric geometry of the two main intrusions; 2) the occurrence of large ultramafic cumulate bodies at the centre of each intrusion; 3) the sub-annular occurrence of differentiates at the borders of each intrusion (Fig. 15a). Accordingly, alkali-basaltic parental magmas of the first intrusion infiltrated and ponded at shallow level in the Precambrian basement (Fig. 15b), leading to crystallization of cumulus olivine that formed the early dunitic body at the centre. Continuous magma supply and efficient diffusion processes resulted in growing of dunitic adcumulates, progressively surrounded by a gabbroic crystal mush whose fractionation produced the felsic differentiates injected as subanular syenitic intrusives or phonolitic dykes. It is important to note that both major element mass balance and PELE modelling of this intrusion necessarily require the removal of a relevant amount (50-60%) of gabbro-diorite cumulates to get syenitic differentiates, a result which apparently contradicts the observed paucity of gabbro-diorite rocks outcropping in the complex (ca. 2%; third column in Supplementary Table 2). This incongruence is overcome in the proposed model (Fig. 15b) assuming that large amounts of gabbroic rocks are hidden in the lower part of the intrusion, whereas the syenitic magmas squeezed out from the mafic crystal mush and emplaced at shallower level of the plutonic body. The entire process could have been favoured by the concomitant effect of the shear failures developed at the tip of the feeding conduit and the progressive compaction of the mafic crystal mush with consequent outward segregation of felsic magmas, as already observed in other shallow levels intrusions (Ratschbacher et al., 2014; Lee et al., 2015). For the second intrusion, it is conceivable that nephelinitic (+ melilite-bearing) parental magmas intruded next to and partly cut the first intrusion, giving rise to extensive ad- to mesocumulitic clinopyroxenites at the centre and a semi-annular fine layered body of ijolite (s.l.) orthocumulates at the periphery; the latter are characterized by a melteigite border at the inner contact with clinopyroxenite and grade to more differentiated ijolites and urtites outward (Fig 15c). Unlike the first intrusion, results from major and trace element modelling are in good agreement with field observation: the amount of clinopyroxenite cumulates and of residual melts required by the model roughly correspond to the relative amounts of clinopyroxenite and ijolite/urtite observed in field, respectively. In any case, like for the first intrusion, the radial injection of magmas may be accounted for by shear failures developed at the tip of the feeding conduit. Carbonatites, representing the last magmatic event of the complex, outcrop at the centre of the clinopyroxenite body, and possibly intruded through the same magma conduit. Noteworthy, the above modelling satisfactorily fits with the main geophysical evidences reviewed by Marangoni and Mantovani (2013) for the Jacupiranga complex, that record the dunite root to 4 km depth and magma conduits of the clinopyroxenite body extending down to ca. 7 km.

## 9. Conclusions

The Jacupiranga complex consists of two main diachronous silicate intrusions: an older dunite-gabbro-syenite body in the NW half part and a younger clinopyroxenite – ijolite (s.l.) body in its SE half part, in turn injected by a carbonatitic core (< 1% vol). Regional geological constraints (i.e. thickness of the neighbouring Mesozoic sedimentary/volcanic cover and estimate of the erosion rate), suggest its shallow emplacement (2 – 3 km depth) between the crystalline basement and the overlying sedimentary units; the ductile rheological behaviour of these sediments likely contributed to confine the uprising magmas hampering their eruption. The shallow emplacement of the complex is also suggested by barometric estimates based on mineral parageneses.

An integrated petrogenetic model based on bulk rock major and trace element analyses and mineral chemistry data allows to conclude, coherently with field observations, that the two silicate intrusions generated from different parental magmas which evolved, under nearly closed system conditions, in two distinct zonally arranged plutonic bodies that grew incrementally from independent feeding systems (Fig. 15). The first intrusion was generated by alkaline to mildly alkaline parental basalts that initially led to the formation of a dunitic adcumulate core, surrounded by gabbroic cumulates, in turn injected by sub-annular syenites intrusives and phonolite dykes.

The second intrusion, formed from nephelinitic parental melts that gave rise to large coarsegrained ad- to meso-cumulitic clinopyroxenites in turn surrounded - and partially cut - by semiannular fine-layered melteigite-ijolite-urtite orthocumulates.

The silicate and carbonatite intrusions have distinctly different Sr-Nd-Pb isotopic compositions supporting the derivation of the relative parental magmas from independent mantle sources. Crustal contamination processes, can not be a priori excluded and are envisaged, particularly for the magmas of the older intrusion that were those firstly injected and stopped within the basement.

A possible origin for Jacupiranga carbonatites at mantle depth is suggested by the absence of carbonatite ocelli in the silicate counterparts of the intrusion, and by fluid and melt inclusions in apatites from carbonatites indicating trapping depths in the range 30-60 km range (Salvioli-Mariani et al., 2012). The alternative hypothesis relating carbonatite to shallow level immiscibility is less plausible, as Jacupiranga carbonatite do not display the LREE/HREE enrichment expected for this process (Brod et al., 2013; Guzmics et al., 2015); in fact, the La/Yb ratio of carbonatite is comparable (or even lower) than that observed in the ijolite *s.l.* rocks. Furthermore, sulphur and iron isotopic data indicate for Jacupiranga carbonatites a primitive nature, thus disfavouring a genesis by shallow level differentiation processes (Johnson et al., 2010; Siquera Gomide et al., 2013).

In a regional framework the acquired isotopic data suggest that gabbro-syenites and carbonatites plausibly derived from independent lithospheric sources that variably recorded long term inherited heterogeneities, whereas the clinopyroxenite-ijolite intrusion displays the involvement of sublithospheric mantle components considered characteristic of the initial plume activity (since 132 Ma) in western Gondwana.

# Acknowledgments

This research started within the joint Italian-Brazilian project on alkaline-carbonatite complexes led by L. Morbidelli (University of Rome) and C.B. Gomes (University of São Paulo). In particular, L.B. acknowledges the important scientific support and assistance of C.B. Gomes, E. Ruberti, L.F. Scheibe during several field works carried out in the eighties. L. Melluso and R. de' Gennaro are gratefully acknowledged for the microprobe analyses obtained at the University of Naples as well as for the fruitful discussion on the observed mineral parageneses. The authors also thank R. Tassinari for ICP-MS analyses at the University of Ferrara, S. Agostini for isotopic analyses at the IGG-CNR of Pisa and P. Sgualdo for his contribution in computations and drawings. Constructive comments provided by the editor F. Pirajno and two anonymous reviewers greatly improved an early version of the paper.

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#### **Figure captions**:

**Figure 1** – Geological sketch map and cross sections of the Jacupiranga complex after Germann et al. (1987) and Gaspar (1989), modified and integrated with data from this work. In the inset are also reported the regional distribution of other Cretaceous alkaline complexes of the Ponta Grossa Arch (modified after Ruberti et al., 2005): BIT, Barra do Itapirapuã; BN, Banhadão; BT, Barra do Teixeira; CN, Cananeia; IP, Ipanema; IT, Itapirapuã; JC, Jacupiranga; JQ, Juquiá; MP, Mato Preto; PAR, Pariquera-Açu; TU, Tunas.

**Figure 2** – a) R1-R2 [R1 = 4Si – 11 (Na + K) – 2 (Fe + Ti); R2 = 6Ca + 2Mg + Al] classification diagram (De La Roche et al., 1980) for the Jacupiranga rocks. Also indicated the Liquid Line of Descent (LLD) calculated by PELE software (Boudreau, 1999) from the least differentiated

compositions of the gabbro-syenite (AG104, AG122, AB126, SD97) and clinopyroxenite-ijolite *s.l.* (ME80, ME82, ANK 111-D) intrusions. Abbreviations: AB – alkali basalt; AG – alkali gabbro; SD – syeno diorite; SY – syenite; P-PH – peralkaline phonolite; DU – dunite; ME – melteigite; IJ – ijolite; UR – urtite; CP – clinopyroxenite. P = calculated parental magma by adding 14-16 wt% of Fo 86-88 olivine to the most basic compositions of the first intrusion.

**Figure 3** – Distribution of Jacupiranga rocks in (a) QAPF diagram (Streckeisen, 1976) reporting the modal composition of the Jacupiranga rocks obtained by mass-balance calculations between whole rock and constituent minerals analyses, and (b) AI (Alkaline Index) vs FSSI (Feldspathoid Silica Saturation Index) diagram (Frost and Frost, 2008). See text for further explanation.

**Figure 4** – Microphotographs (plane polarized light) of rocks of the Jacupiranga north-western intrusion: (a) serpentinized dunite, (b, c) alkali-gabbro, (d) syeno-diorite, (e) syenite, (f) phonolite. Mineral abbreviations as in Table 2.

**Figure 5** – Microphotographs (plane polarized light) of rocks of the Jacupiranga south-eastern intrusion: (a) clinopyroxenite, (b) melteigite, (c) ijolite, (d) urtite, (e) ankaratrite. In (f) SEM micrograph showing the nepheline-melilite-kalsilite-clinopyroxene mineralogical association in melteigite. Mineral abbreviations as in Table 2.

**Figure 6** – Diopside-nepheline-akermanite-silica systems (after Onuma and Yamamoto, 1976). The normative compositions of the two intrusions of Jacupiranga plot on opposite sides of the diopside-nepheline join, due to the ubiquitous presence of normative Ca-orthosilicate (Ca<sub>2</sub>Si<sub>2</sub>O<sub>6</sub>, a virtual indicator of akermanite melilite) in the extremely silica-undersaturated rocks of the second intrusion.

**Figure 7** – Cumulative curves of crystallizing minerals predicted by PELE software starting from the least differentiated compositions of the gabbro-syenite intrusion (AG104, AG122, AB126, SD97) assuming 0.1 GPa pressure and  $fO_2$  between QFM+1 and QFM+2.

**Figure 8** – (a) Chondrite (Ch)-normalized REE and (b) primordial mantle (PM)-normalized incompatible element distributions for the Jacupiranga rocks of the first intrusion (dunite-gabbrosyenite). Normalizing factors from Sun and McDonough (1989). Abbreviations as in Fig. 2.

**Figure 9** – Primordial mantle (PM)-normalized incompatible element distributions from selected alkali-gabbros, syenites and peralkaline phonolite of the first intrusion. The trace element fractionation modelling performed by PELE software (Boudreau, 1999) satisfactorily fits the residual compositions of felsic differentiated (Sy JC119, P-Ph JC121-D) from alkali-gabbro JC104. Normalizing factors from Sun and McDonough (1989).

**Figure 10** – Cumulative curves of crystallizing minerals predicted by PELE software (Boudreau, 1999) starting from the least differentiated compositions of the clinopyroxenite-ijolite *s.l.* intrusion (ME80, ME82, ANK 111-D) assuming 0.1 GPa pressure and  $fO_2$  QFM+2.

**Figure 11** – (a) Chondrite (Ch)-normalized REE and (b) primordial mantle (PM)-normalized incompatible element distributions for the Jacupiranga rocks of the second intrusion (clinopyroxenite-ijolite *s.l.*). For comparison, the composition of a mafic dyke from Jacupiranga is also reported after Menezes et al. (2015). Normalizing factors from Sun and McDonough (1989). Abbreviations as in Fig. 2.

**Figure 12** – Primordial mantle (PM)-normalized incompatible element distributions from selected melteigite, urtites and clinopyroxenites of the second intrusion. The trace element fractionation

modelling performed by PELE software (Boudreau, 1999) satisfactorily fits the residual compositions of felsic differentiates (Urtite JC74) from ankaratrite JC111-D. Normalizing factors from Sun and McDonough (1989).

**Figure 13** – (a) Sr-Nd and (b) Pb isotopic composition of the Jacupiranga rocks compared with data from Gough and Tristan hot spot tracks (Hoernle et al., 2015), Paraná-Etendeka CFB and Atlantic MORB 10°S-40°S (after Rocha-Junior et al., 2013; Hoernle et al., 2015 and references therein). Smaller symbols for Jacupiranga rocks refer to data by Huang et al., 1995. Reference mantle endmembers (DM, EMI, EMII and HIMU) are also reported for comparison (Stracke, 2012).

Figure 14 –  $\delta^{13}$ C (‰) vs C (wt %) diagram for Jacupiranga rocks showing a complete decoupling between carbonatites and the associated silicate rocks. By contrast, the Juquià complex displays a gradual compositional variation from silicate rocks to carbonatites, in parallel with the increase of observed carbonatitic ocelli in essexites and ne-syenites. For comparison  $\delta^{13}$ C values of Brazilian carbonatites are reported from Santos and Clayton (1995).

Figure 15 – Cartoon illustrating the inferred mode of emplacement of the Jacupiranga complex; a) Model of magma intrusion, propagation and cup-shaped cumulate growth (modified after the analogue experiment of Mathieu et al., 2008); b) refers to the first dunite-gabbro-syenite intrusion where a continuous infiltration of alkali-basaltic parental magmas leads to early cumulus dunites at the centre of the magmatic body and, subsequently, to gabbroic-syenitic differentiates emplaced with subanular/inverted cone geometry; c) refers to the second clinopyroxenite-ijolite *s.l.* intrusion partly cutting the southern portion of the first intrusion; in this case, the nephelinitic parental magmas give rise to clinopyroxenitic cumulates constituting the majority of the intrusion and a cup-shaped layered body of ijolite-melteigite-urtite intrusives. In turn, the carbonatite body intrudes the

clinopyroxenite cumulates possibly trough the same feeding system. Arrows indicate magma flow directions. Rock abbreviations as in Table 1. Colours and symbols as in Fig. 1.

## **Supplementary Figure captions:**

**Supplementary Figure 1** – Sampling location used for mapping of the Jacupiranga complex (Jc).

**Supplementary Figure 2** – Wo-En-Fs (mol. %) ternary diagram for pyroxenes and amphiboles from Jacupiranga intrusions. Also reported Fo (mol. %) compositional range for olivines. Tie dashed lines join the clinopyroxene-amphibole-olivine compositions in the same sample. Abbreviations: Wo – wollastonite; Di – diopside; En – enstatite; Fs – ferrosilite; Fo – forsterite; Fa – fayalite. Rock abbreviations as in Table 1.

**Supplementary Figure 3** – Or-Ab-An (mol. %) ternary diagram for feldspars from Jacupiranga Gabbro-Syenite intrusion. Abbreviations: Or – orthoclase; Ab – Albite; An – anorthite. Rock abbreviations as in Table 1.

**Supplementary Figure 4** – Al-Mg-Fe+Mn (a.p.f.u. %) ternary diagram for micas from Jacupiranga intrusions. Rock abbreviations as in Table 1.

Supplementary Figure 5 –  $Mg/(Mg+Fe^{2+})$  vs Si (a.p.f.u.) classificative diagram for amphiboles from Jacupiranga intrusions. Rock abbreviations as in Table 1.

# **Table captions:**

**Table 1** – Bulk rock major and trace element composition of Jacupiranga complex.

#### Footnote

Abbreviations: AG = Alkali-Gabbro; AB = Alkali-Basalt; SD = Syeno-Diorite; Sy = Syenite; P-Ph = Peralkaline phonolite; Du = Dunite; Ij = Ijolite; Me = Melteigite; Ur = Urtite; Cp = Clinopyroxenite; Carb = Carbonatite; \* = Melilite bearing; LOI = Loss on Ignition; nd = not detected; Mg# = mol [MgO/(MgO+FeO)] with Fe<sub>2</sub>O<sub>3</sub>/FeO = 0.15

(1) XRF Major and trace element analyses already published (Courtesy L. Beccaluva) in Comin-Chiaramonti and Gomes, 2005.

# **Table 2** – Mineral paragenesis of Jacupiranga rocks.

#### Footnote

Abbreviations: ol – olivine; chr – Cr-spinel; cpx – clinopyroxene; phl/bt – phlogopite/biotite; amph – amphibole; mel – melilite; mt – magnetite; ilm – ilmenite; prv – perovskite; ttn – titanite; neph – nepheline; kls – kalsilite; nos/sdl – nosean/sodalite; pl – plagioclase; af – alkali feldspar; qz = quartz; ap/bri – apatite/brianite; gt – garnet; carb – carbonates; sulf – sulfides; badd – baddeleite; chev – chevkinite; zrc – zircon; bar – barite; wohl – wohlerite; thu – thorurite; fluo – fluorite; thor – thorite; stron – stronalsite; cels – celsian; bar – barite; pcl – pyrochlore; pbc – Pb-carbonate. Rock abbreviations as in Table 1. Brackets refer to not ubiquitous phases.

**Table 3** – Average modal composition (wt%) and range (in brackets) of Jacupiranga rocks inferred by major element mass balance calculations between whole rocks and constituent minerals. Rock abbreviations as in Table 1. Mineral abbreviations as in Table 2.

**Table 4** – Sr-Nd-Pb isotopic composition of Jacupiranga rocks. Rock abbreviations as in Table 1.

## Footnote

<sub>Init</sub> = initial values calculated at 130 Ma

**Table 5**– Elemental (wt %) and isotopic ( $\delta^{13}$ C %) carbon composition for Jacupiranga rocks. New analyses for selected silicate and carbonatite rocks from the Juquia complex are also reported for comparison (samples from Beccaluva et al., 1992). Abbreviations as in Table 1. See text for further explanation.

## **Supplementary Table caption:**

**Supplementary Table 1:** Representative analyses of constituent minerals from Jacupiranga rocks. Abbreviations for rocks and minerals as in Table 1 and 2. (1) = analysis carried out at the Laboratories of Modena University. Other analyses carried out at Naples University.

**Supplementary Table 2:** Synthesis of results from different (independent) petrogenetic models simulating the magma differentiation processes of Jacupiranga silicate intrusions. Field estimates on the extent of distinct lithologies are also reported for comparison.

Footnotes: 1- olivine composition of dunite cumulates used to infer the theoretical primary magma; 2- olivine composition observed in alkali-gabbros used in mass balance calculations; - = not required by the model; in brackets the F value estimated from PELE incompatible element best fit. Abbreviations for rocks and minerals as in Table 1 and 2.