



Nitrogen and argon in Sung Valley and Ambadongar carbonatite complexes: Evidence of incomplete homogenization of mantle and recycled components



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ABSTRACT

Nitrogen and argon concentrations and isotopic ratios have been measured in mineral separates (carbonates, apatites and magnetites) of two temporally and spatially different Indian carbonatites from Sung Valley (107 Ma) and Ambadongar (65 Ma). Gases were extracted by vacuum crushing (carbonates, apatites and magnetites) and stepwise pyrolysis (only apatite) to look for the trapped gases and resolve the different components. $\delta^{15}\text{N}$ varies from +1.4‰ to +13.6‰ (with accompanying $^{40}\text{Ar}/^{36}\text{Ar}$ varying from 900 to 4956) in Sung Valley, and from –3.4 to +14.2 (with accompanying $^{40}\text{Ar}/^{36}\text{Ar}$ varying from 353 to 5301) in Ambadongar. This clearly shows the presence of more than one component in these carbonatites. We identify primary and recycled nitrogen and argon in these carbonatites. Additionally, N may be present in other speciation, like solid inorganic form with elevated $\delta^{15}\text{N}$ ratios up to 16.9‰. Unlike the well-studied carbonatites from Kola, these Indian carbonatites do not show complete homogenization of the different components that can be hosted in different populations of inclusions and, as a result, can be identified. Preservation of such heterogeneity indicates that carbonatite magmatism originated from a small-scale heterogeneous subcontinental mantle initiated during lithospheric rifting.

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1. Introduction

Carbonatites are magmatic rocks with high modal abundance (>50 wt%) of carbonate minerals such as calcite, dolomite, ankerite and siderite, along with Sr, Ba, P and the light rare-earth elements (LREE) (Nelson et al., 1988). While carbon is present in the mantle only in trace amounts (~16–300 ppm; Dasgupta and Hirschmann, 2010), the source of the huge amount of carbon in carbonatites has always been a matter of debate. In these rocks, carbon is believed to be either juvenile, based on excess ^{129}Xe (Sasada et al., 1997), or recycled, based on enriched $\delta^{13}\text{C}$ signatures (Ray et al., 1999, 2000). However, $\delta^{13}\text{C}$ also depends on several other parameters like the $\text{CO}_2/\text{H}_2\text{O}$ ratio, the C-bearing species in fluids (i.e. H_2CO_3 or HCO_3^-), and the degree of equilibrium or Rayleigh fractionation (Reid and Cooper, 1992).

Contribution from recycled component in carbonatites can be better resolved by the means of nitrogen isotopes, because the nitrogen isotopic ratios of air, the mantle and sediments are distinctly different (Haendel et al., 1986; Marty and Humbert, 1997;

Mohapatra and Murty, 2004). As compared to the well mixed atmospheric reservoir ($\delta^{15}\text{N} \equiv 0$), crustal nitrogen has ^{15}N -enriched isotopic signatures with $\delta^{15}\text{N}$ varying from +2‰ (Busigny et al., 2003) up to +15‰ (Haendel et al., 1986). The $\delta^{15}\text{N}$ of the degassed MORB mantle varies from –15‰ to –5‰ (Marty and Humbert, 1997; Mohapatra and Murty, 2000, 2004). The nitrogen isotopic signature of carbonatites from Oldoinyo Lengai, which is associated with the East African Rift, is MORB-like (Fischer et al., 2009). On the other hand, the source of the Kola carbonatites, believed to be located in the deep mantle based on noble gases (Marty et al., 1998; Tolstikhin et al., 2002), have a heavy nitrogen composition of +1‰ to +3‰, which has been attributed to subduction, or interaction with meteoric water (Dauphas and Marty, 1999; Buikin et al., 2014).

Unlike the carbonatites from Kola, noble gas ratios of $^3\text{He}/^4\text{He}$, $^{21}\text{Ne}/^{22}\text{Ne}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ in the Indian carbonatites indicate shallower mantle sources with contribution from MORB-like sources (Basu and Murty, 2006; Murty et al., 2007). So far, nitrogen isotopic data from carbonatites are very scarce because of experimental difficulties in measuring the very low concentration of primary gases as compared to the amount of atmospheric and organic components derived from contamination processes. The present effort is the first extensive study of nitrogen from Indian carbonatites. In

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this study, we investigate the source of nitrogen in the carbonatites from Sung Valley and Ambadongar to characterize the role of subduction in carbonatitic magmatism. Earlier, studies from diamonds have shown that positive $\delta^{15}\text{N}$ are often associated with light carbon, which indicates subduction of both carbon and nitrogen simultaneously with crustal material (Cartigny et al., 1998; Mohapatra and Honda, 2006). Thus, identification of subducted nitrogen in carbonatites could be an indicator of subducted carbon as well.

In addition to nitrogen isotopic composition, the N/Ar elemental ratio is also very useful to track any recycled nitrogen introduced into the mantle during subduction. The electronic configuration of molecular nitrogen makes it chemically inert like noble gases. Its radius (75 pm) is similar to that of Ar (71 pm), and they have been found to have comparable behavior during magma generation and degassing (Marty and Humbert, 1997). But, Ar is not chemically bound to the silicate network as nitrogen in the form of NH_4^+ ions, and so is less efficiently recycled than nitrogen to the mantle. The $\text{N}_2/^{36}\text{Ar}$ has been constrained to be 5×10^6 for the shallow mantle and 3×10^5 for the deep mantle source of Kola, where the elemental ratios are consistent with mixing between air, mantle and recycled sediments (Dauphas and Marty, 1999). Here we discuss our nitrogen results for the first time, in conjunction with simultaneously obtained argon results. The argon results have been reported in Basu and Murty (2006). The locations of the studied carbonatites are shown in Fig. 1. Some details of their ages and previous isotopic works are discussed below.

2. Geological settings and earlier studies

2.1. Sung Valley

The Sung Valley carbonatite–alkaline province, associated with the Rajmahal-Bengal-Sylhet Traps occurs as dykes or sheet-like bodies intruding into the alkaline rocks of the Assam-Meghalaya Plateau (Kumar et al., 1996). The carbonatitic complex is bounded by the Brahmaputra fault to the north and the Dauki fault to the south (Kumar et al., 1996). Their ^{40}Ar – ^{39}Ar age of 107 Ma, together with the spatial proximity to the Sylhet traps, are consistent with the hypothesis of their Kerguelen plume origin (Ray et al., 1999). The carbonatites are mainly composed of calcite with accessory dolomite, siderite, apatite, phlogopite, pyrochlore, zircon and magnetite. The $\delta^{18}\text{O}$ (+7.1‰ to +8.4‰) is consistent with expected primary carbonatite melt composition whereas the $\delta^{13}\text{C}$ (–3.2‰) indicates incorporation of recycled inorganic carbon from subducted oceanic crust into the subcontinental lithospheric mantle (Ray et al., 1999; Ray and Ramesh, 2006). The initial ϵ_{Sr} (+5.3 to +7.8) and ϵ_{Nd} (+1.7 to +2.3) values suggest an enriched mantle source (Veena et al., 1998). The noble gas ratios $^{20}\text{Ne}/^{22}\text{Ne}$ (up to 12.1) and $^{129}\text{Xe}/^{132}\text{Xe}$ (up to 1.06), suggest presence of mantle gases (Basu and Murty, 2006). The $^3\text{He}/^4\text{He}$ (up to 9.01 R/R_A) and $^{21}\text{Ne}/^{22}\text{Ne}$ (up to 0.069) ratios are consistent with the presence of MORB-like gases (Basu and Murty, 2006).

2.2. Ambadongar

The Ambadongar carbonatite–alkaline complex, associated with the Deccan Traps, is located within the Narmada-Son rift zone in Western India and has a ^{40}Ar – ^{39}Ar age of 65 Ma (Ray et al., 2000). Late stage ferrocarnatites occur as small plugs within early stage calcitic carbonatites. For this study, samples have been analyzed from the calcitic carbonatite, which contains apatite, magnetite, barite, phlogopite, aegerine, zirconite and pyrochlore. Ambadongar carbonatites have been derived from two parent magmas and, model-based initial $\delta^{13}\text{C}$ (–5.5‰ to –3.4‰) and

$\delta^{18}\text{O}$ (+7.5‰ to 9.5‰) composition of the magmas are consistent with a mantle source (Ray et al., 2000; Ray and Ramesh, 2006). The high, ^{13}C -enriched $\delta^{13}\text{C}$ (\sim –3.4‰) of the group-II magma can be attributed to contribution from ^{13}C -enriched, recycled crustal carbon (Ray et al., 2000). The initial ϵ_{Sr} (+15.1 to +19.2) and ϵ_{Nd} (+1.7 to +2.3) values for the calcitic carbonatites suggest an enriched mantle source (Simonetti et al., 1998). These isotopic ratios are comparable to that of the proximal Deccan flood basalts of similar age. Therefore, the Reunion hot spot responsible for the Deccan flood basalts may also have contributed to the generation of the parental melt of the Ambadongar carbonatite (Simonetti et al., 1998). The noble gas ratios $^{20}\text{Ne}/^{22}\text{Ne}$ (up to 10.2), $^{21}\text{Ne}/^{22}\text{Ne}$ (up to 0.054), and $^{129}\text{Xe}/^{132}\text{Xe}$ (up to 1.06), clearly show the presence of mantle gases, consistent with MORB-like component admixed with crustal noble gases (Basu and Murty, 2006).

3. Samples and experimental methods

3.1. Sample characterization

For this study, we opted for coarse-grained calcitic carbonatites. We discarded the ferrocarnatites as they are often affected by post magmatic alteration processes induced by late stage hydrothermal fluid circulation. Such processes can enrich the samples in atmospheric gases. The dark brown appearances of the iron-rich ferrocarnatites contrast markedly from the more common white calcitic carbonatites, and the carbonates are often oxidized. We separated mm-sized grains of calcites (Sung Valley and Ambadongar), apatites (Sung Valley) and magnetites (Sung Valley) by hand picking under optical microscope. XRD on the mineral separates was done to confirm mineral homogeneity.

Thin section studies also aided in the correct selection of samples. Very often, oxidizing Fe-rich fluids have penetrated along grain boundaries, progressively introducing Fe^{3+} into the original carbonate minerals. We discarded any samples marked by iron oxide staining on the calcite or dolomite (Fig. A.1). Early formed magmatic calcite and dolomite accompanied by silicates and oxides tend to react with water at low temperature to form quartz, zeolites, fluorite, chlorite, rutile and haematite. In order to avoid late stage carbonatites affected by water with dissolved atmospheric gases that can dilute the mantle signatures, we discarded samples containing any of the above late stage minerals.

Apatites in carbonatites commonly occur as fluorapatite ($\text{Ca}_{10}(\text{PO}_4)_6\text{F}_2$) and persist from early to late stage carbonatites. Various trace elements (Ba, Sr, Na, REEs) can be used to distinguish between early and late stage apatites that may be formed by secondary processes (Hogarth, 1989). Based on the trace element analyses of Ba, Sr, Na, REEs in some of the apatites, there is no indication that they are of secondary origin, or have been generated in the later stages of carbonatitic magmatism (Table A.1).

3.2. Gas extraction and analysis

Vacuum crushing (VC) of mineral separates (1–2 g) preferentially releases trapped gases from fluid inclusions with minimum damage to lattice structure. A crusher with a magnetic piston activated by external solenoids was employed for the gas extraction. VC was done sequentially using 15, 300 and 300 strokes for carbonates and 15, 500 and 500 strokes for apatites and magnetites to progressively release gases from smaller fluid inclusions. In most cases, maximum release of gases occurred in the second step of crushing. Apatites (\approx 500 mg) have been also analyzed by step-wise pyrolysis (P) to release total gases present in the samples (fluid inclusions + lattice structure), and to separate the different components by their release at different temperatures. An initial

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