



N-isotope composition of the primitive mantle compared to diamonds

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ARTICLE INFO

Article history:

Received 28 July 2014

Accepted 10 February 2015

Available online 23 February 2015

Keywords:

Nitrogen

Isotopes

Primitive mantle

Crust

Recycle

Neoproterozoic

ABSTRACT

The nitrogen isotopic composition of the Earth's primitive mantle is controversial. Chromium-isotopic ratios of various terrestrial minerals and rocks, and chondritic meteorites are consistent with the silicate Earth being a mixture of enstatite and carbonaceous chondrites. From their relative proportions and N-isotope compositions we estimate that the bulk primitive mantle $\delta^{15}\text{N}$ is $-7 \pm 3\%$. The negative value, as also evidenced by mantle-derived oceanic basalts and diamonds, is an intrinsic long-term feature of Earth's mantle. Some enstatite chondrite-like $\delta^{15}\text{N}$ values down to -24% measured in very rare diamonds could be interpreted as a heterogeneous mantle. $\delta^{15}\text{N}$ values in oceanic island basalts derived from the deep mantle have three components: deep mantle of $\sim -9\%$ consistent with estimate, recycled sediments of about 15% , and atmospheric N incorporated from groundwater and/or subducted atmospheric N (Mohapatra and Murty, 2000a; Chem. Geol. 164, 305–320). Some enriched $\delta^{15}\text{N}$ values in MORB and OIB result from degassing fractionation. Shift of the upper mantle from an initial -7% to -5% by the Neoproterozoic can be explained by a combination of sediment recycling through subduction and upper mantle magma degassing processes.

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1. Introduction

The origin and evolution of nitrogen in the mantle have been controversial since the 1950's (Brown, 1953; Pepin and Porcelli, 2002). The isotopic composition of nitrogen shows large different variations in extraterrestrial samples (e.g., chondritic meteorites; Clayton, 1981; Wänke, 1981), which makes this element a useful tracer of mass exchange between the surface and deep-Earth reservoirs and of fluid/melt-rock interactions in the crust and mantle (Busigny and Bebout, 2013; Busigny et al., 2011; Cartigny, 2005; Cartigny and Marty, 2013; Halama et al., 2012; Philippot et al., 2007), and a potentially important tracer for the origin of the terrestrial silicates and volatiles (Busigny et al., 2011; Cartigny, 2005; Cartigny and Marty, 2013; Jia and Kerrich, 2004; Kerrich et al., 2006; Marty, 2012). Nitrogen isotopic composition is expressed by the $\delta^{15}\text{N}$ parameter defined as $\delta^{15}\text{N} = [({}^{15}\text{N}/{}^{14}\text{N})_{\text{sample}}/({}^{15}\text{N}/{}^{14}\text{N})_{\text{standard}} - 1] \times 1000$, where the standard is the atmospheric N_2 . Enstatite (E) chondrites have a total range of $\delta^{15}\text{N}$ from -15% to -43% , with most values between -20% and -30% (Inger and Kaplan, 1974; Grady et al., 1986; Kung and Clayton, 1978). In contrast, carbonaceous (C) chondrites have a range from $+16\%$ to $+52\%$ (Fig. 1; Inger and Kaplan, 1974; Kerridge, 1985; Kung and Clayton, 1978; Lewis et al., 1983; Robert and Epstein, 1982).

An enstatite (E) chondrite model for the Earth has been proposed by several workers (Javoy, 1997, 1998; Tolstik and Marty, 1998) based on two principal lines of evidence: similarity of oxygen isotope compositions of the Earth–Moon system with E-chondrites (Javoy and Pineau, 1983; Javoy et al., 1986), and rare ${}^{15}\text{N}$ -depleted diamonds down to -21% (Shatskii et al., 2011) or -24% (Cartigny, 2005; Cartigny et al., 1997, 1998). The most depleted nitrogen isotope compositions of -40% are also found in a few diamonds (Palot et al., 2012), although both the size and location of this reservoir are unknown. According to Javoy (1998), 99.8% of the planet is of E-chondrite composition, contributing to the nitrogen budget of the Upper Earth (UE: upper mantle + crust + ocean–atmosphere) after upper–lower mantle stratification. A late veneer of C1 chondrite and/or comets of 0.14% Earth mass brought in 30% of the UE budget. In this model, mantle $\delta^{15}\text{N}$ would have evolved from an initial state characterized by E-chondrite type material having $\delta^{15}\text{N}$ values of less than -25% , matching those of some rare diamonds (Cartigny, 2005; Cartigny et al., 1997, 1998; Palot et al., 2012; Shatskii et al., 2011) to about -5% by the Archean (Javoy, 1997, 1998). This model was questioned based on no direct evidence or record of a secular change in mantle $\delta^{15}\text{N}$ values (Cartigny and Marty, 2013). They rather propose that the marked nitrogen isotopic contrast between the mantle and the Earth's surface could reflect origins of the nitrogen disequilibrium. This proposal might provide insights into mantle–surface interactions over geological time including recycling of surface sediments into the deep mantle, although the cause of such disequilibrium is not fully understood (Cartigny and Marty, 2013).

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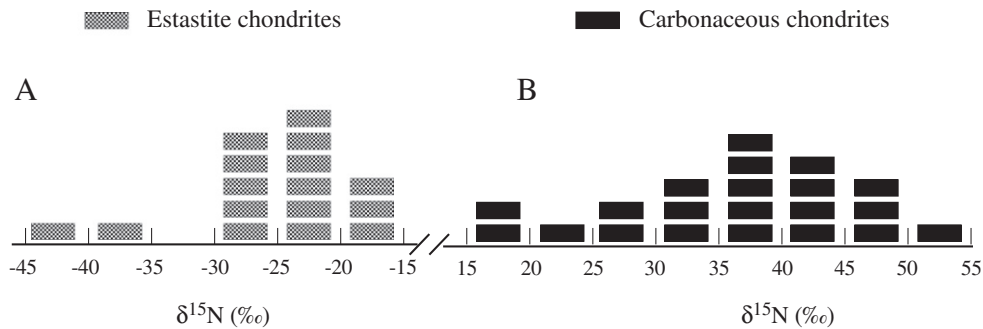


Fig. 1. $\delta^{15}\text{N}$ values for enstatite chondrites (A) and carbonaceous chondrites (B). Enstatite chondrites from Injerd and Kaplan (1974), Kung and Clayton (1978), and Grady et al. (1986). Carbonaceous chondrites from Injerd and Kaplan (1974), Kung and Clayton (1978), Robert and Epstein (1982), and Kerridge (1985).

Tolstikin and Marty (1998) also suggested that the Earth accreted from E-chondritic material having a $\delta^{15}\text{N}$ of -30‰ . In their model, due to a major phase of accretion, impact(s), core segregation, and mantle layering, the upper mantle underwent extensive degassing from 4.9×10^{23} mol N at 4.5 Ga, at ~ 60 Ma post accretion, to 2×10^{17} mol at 4.36 Ga accompanied by a shift of $+30\text{‰}$ stemming from hydrodynamic fractionation in the upper atmosphere. That shift generates the present value of -5‰ observed in mid-ocean ridge basalts (cf. Marty and Humbert, 1997). Their model also leads to an atmospheric $\delta^{15}\text{N}$ of $+2.5\text{‰}$ at 4.3 Ga, and further degassing of mantle N $\sim -5\text{‰}$ allowed this element to reach its present-day value of 0‰ .

A pure E-chondritic mantle hypothesis has been questioned based on various lines of evidence: (1) the chemical (Si/Mg) and isotopic ($^{187}\text{Os}/^{186}\text{Os}$) compositions of the bulk earth are not consistent with enstatite chondrites or C1 chondrites alone (Allègre et al., 1995a; Drake and Righter, 2002); and (2) modern plumes, such as the Icelandic plume, which include a deep mantle component as indicated by noble gas isotopes and Nb/Th ratios (Breddam et al., 2000; Moreira et al., 2001), do not have $\delta^{15}\text{N}$ of -25‰ ; rather they are characterized by $\delta^{15}\text{N}$ of -10.5‰ to -0.5‰ (Marty et al., 1991).

More recently, Marty and Dauphas (2003) proposed a new sediment-recycling model for the isotopic composition of nitrogen in different mantle reservoirs. Nitrogen in the upper mantle, as sampled by mid-oceanic ridge basalts and diamonds having a mean $\delta^{15}\text{N}$ of -5‰ , originated from recycling of ^{15}N -depleted Archean chert-like N into the upper mantle, whereas nitrogen in the lower mantle, as sampled by modern mantle plumes such as ocean island basalts (OIB) with $\delta^{15}\text{N}$ of 1 to 8‰ , was derived from recycling of ^{15}N -enriched post-Archean sediments into the lower mantle.

The recycling model developed by Marty and Dauphas (2003) relies heavily both on selected data for Archean cherts of Beaumont and Robert (1999), and on a sequence of assumptions: (1) that organic N in cherts is representative of recycled Archean sediment/crust; and (2) that there has been a switch from recycling of sediments into upper mantle in the Archean, but into the lower mantle post-Archean. A curious feature of their model is an unspecified primitive mantle $\delta^{15}\text{N}$ (see Fig. 3 of Marty and Dauphas (2003)). This is in contrast to initial ratios for Sr, Nd, Hf, Pb, and Os isotopes, which are used to constrain models of mantle evolution (Dickin, 1997). Hence their model was seriously questioned by Cartigny and Ader (2003) and Kerrich and Jia (2004).

Recent studies on Cr-isotopes ($^{53}\text{Cr}/^{52}\text{Cr}$) of various terrestrial and extraterrestrial geological samples show great potential for an independent method to estimate the nitrogen isotope composition of the primitive mantle. The radionuclide ^{53}Mn is unstable, decaying to ^{53}Cr by beta emission with a half-life of 3.7 ± 0.4 Ma (Honda and Imamura, 1971). Given that the Cr-isotope composition of the bulk Earth, E-, and C1-chondrites are known (Lugmair and Shukolyukov, 1998; Shukolyukov and Lugmair, 1998), it is possible to conduct a mass balance for N and N-isotopes.

Consequently, the aims of this paper are: (1) to re-examine published mantle N-isotope and concentration data recorded by diamonds, mid-oceanic ridge basalts, oceanic island basalts, and mafic alkaline magmas such as lamproites; (2) to discuss the differences between them; (3) to estimate the $\delta^{15}\text{N}$ of the primitive mantle utilizing mass balance calculations based on the Cr-isotope data; and (4) to discuss possible evolution of mantle $\delta^{15}\text{N}$ based on mass balance calculations.

2. N-isotope characteristics of the upper mantle

The nitrogen isotope composition of the upper mantle has been inferred from two sets of geological samples: mid-ocean ridge basalts (MORBs) and diamonds. Fig. 2 depicts a worldwide compilation of N-isotopic compositions of upper mantle-derived material, where initial results on fibrous diamonds in Zaire were reported by Javoy et al. (1984) and Boyd et al. (1987, 1992). The $\delta^{15}\text{N}$ in fibrous diamonds show a very tight range of -8.7 to -1.7‰ , with a mean of -5‰ . Eclogitic diamonds have a mean $\delta^{15}\text{N}$ value of $-5.5 \pm 2.0\text{‰}$ ($n = 40$), nearly identical to that of fibrous diamonds (Cartigny, 2005; Cartigny et al., 1998). The mean $\delta^{15}\text{N}$ value of peridotitic diamonds is also negative ($-8 \pm 5.2\text{‰}$; $n = 42$), although the range of this type of diamonds is larger than fibrous counterparts (see Fig. 2; Cartigny et al. (1997, 2009) and references therein). The diamonds are thought to grow in the upper mantle from carbonate melts reacting with continental lithospheric mantle (CLM), and are mostly Archean in age (Richardson et al., 1984; Richardson et al., 2001).

The N-isotope data on mid-ocean ridge basalts having $^{40}\text{Ar}/^{36}\text{Ar} > 1000$ also show ^{15}N -depleted values between -8.1 and -0.5‰ , with a median of $-3.4 \pm 1.6\text{‰}$. Samples with very high $^{40}\text{Ar}/^{36}\text{Ar}$ exhibit $\delta^{15}\text{N}$ close to -5‰ (Fig. 2; Marty and Humbert, 1997; Marty and Zimmermann, 1999). For some mid-ocean ridge basalt samples showing a relatively positive $\delta^{15}\text{N}$ values, they generally associated with low $^{40}\text{Ar}/^{36}\text{Ar}$ ratios, which might reflect contamination by surface material or source heterogeneity (Cartigny and Marty, 2013). Collectively, the $\delta^{15}\text{N}$ signature of MORB and diamonds are comparable, albeit with MORB extending to less negative values. According to Marty and Zimmermann (1999), the difference is attributed to distinct mantle sources for diamond forming and MORB magmas: diamond is thought to represent the nitrogen isotopic ratio of deeper asthenospheric mantle liquids, whereas the latter is characterized by shallow-level mantle N. However, Marty and Dauphas (2003) conclude that the MORB and diamond sample common volatile sources after correction for fractional degassing and fractionation during diamond growth respectively. Alternatively, the ^{15}N -depleted isotopic composition of both diamond and MORB is a long-term feature of the Earth's upper mantle (Cartigny et al., 1997, 1998), and the relatively higher or positive $\delta^{15}\text{N}$ values in some MORB samples compared with diamonds is attributed to nitrogen isotope fractionation during shallow degassing (Cartigny and Ader, 2003).

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