



# The $^{15}\text{N}$ -enrichment in dark clouds and Solar System objects

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## ABSTRACT

The line intensities of the fundamental rotational transitions of  $\text{H}^{13}\text{CN}$  and  $\text{HC}^{15}\text{N}$  were observed towards two prestellar cores, L183 and L1544, and lead to molecular isotopic ratios  $140 \leq ^{14}\text{N}/^{15}\text{N} \leq 250$  and  $140 \leq ^{14}\text{N}/^{15}\text{N} \leq 360$ , respectively. The range of values reflect genuine spatial variations within the cores. A comprehensive analysis of the available measurements of the nitrogen isotopic ratio in prestellar cores show that molecules carrying the nitrile functional group appear to be systematically  $^{15}\text{N}$ -enriched compared to those carrying the amine functional group. A chemical origin for the differential  $^{15}\text{N}$ -enhancement between nitrile- and amine-bearing interstellar molecules is proposed. This sheds new light on several observations of Solar System objects: (i) the similar N isotopic fractionation in Jupiter's  $\text{NH}_3$  and solar wind  $\text{N}^+$ ; (ii) the  $^{15}\text{N}$ -enrichments in cometary HCN and CN (that might represent a direct interstellar inheritance); and (iii)  $^{15}\text{N}$ -enrichments observed in organics in primitive cosmomaterials. The large variations in the isotopic composition of N-bearing molecules in Solar System objects might then simply reflect the different interstellar N reservoirs from which they are originating.

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## 1. Introduction

Nitrogen, the fifth most abundant element in the Universe, exists naturally as a highly volatile gas ( $\text{N}_2$ , N) and a mixture of compounds of varying volatility (such as  $\text{NH}_3$ , HCN, and HNC). The relative abundances and isotopic compositions of these different nitrogen occurrences in various astronomical sources can provide useful clues to the origin and history of the Solar System.

The Sun formed from a cold and dense core embedded in its parental interstellar molecular cloud rich in gas and dust. The so-called "protosolar nebula" (PSN) is the evolutionary stage issued from the collapsing prestellar core. The nitrogen volatile isotopologues in this nebula may have been fractionated with respect to the original interstellar material, i.e. the isotopic ratio measured in these molecules may differ from the elemental ratio. Such fractionation processes are invoked to explain the large enhancements of the D/H ratio measured in several molecular species in prestellar cores (e.g. Caselli et al., 2003; Roueff et al., 2005). The efficiency of these processes however depends on the physical conditions in the core during its collapse (Flower et al., 2006). One of the current challenges in astrochemistry is to follow the chemical composition of a starless core during its evolution towards a planetary system. The related challenge in cosmochemistry is to identify, in primitive objects of the Solar System, residual materials from the original cloud.

The Sun is the largest reservoir of nitrogen in the Solar System. Isotopic measurements of solar wind trapped in lunar soils

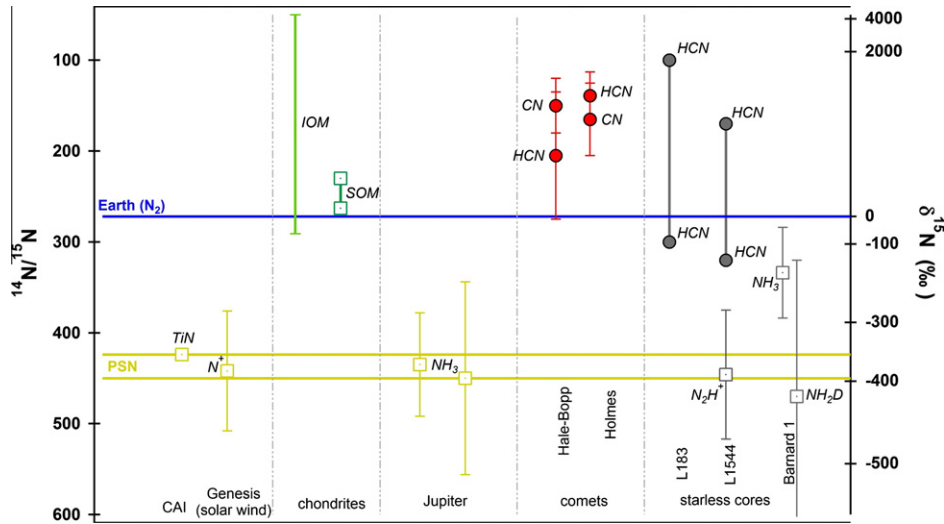
(Hashizume et al., 2000), analysis of Jupiter's atmosphere (Fouchet et al., 2000; Owen et al., 2001) and osbornite (TiN), considered as the first solid N-bearing phase to form in the cooling protosolar nebula (Meibom et al., 2007), all independently showed that nitrogen in the PSN was much poorer in  $^{15}\text{N}$  than the terrestrial atmosphere. The analysis of the present-day solar wind trapped on Genesis targets finally concluded on and confirmed these previous studies. The solar wind is depleted in  $^{15}\text{N}$  relative to inner planets and meteorites, and define the following atomic composition for the present-day Sun  $^{14}\text{N}/^{15}\text{N} = 441 \pm 5$  (Marty et al., 2010, 2011). The isotopic composition of nitrogen in the outer convective zone of the Sun has not changed through time and is considered as representative of the PSN. In the present paper, we only consider the original/primary N isotopic fractionation, as opposed to secondary  $^{15}\text{N}$ -enrichments acquired through atmospheric process (e.g. Titan, Mars) for example. In the remainder of the paper and for the sake of clarity, the elemental isotopic ratio is noted  $^{14}\text{N}/^{15}\text{N}$ , whilst the isotopic ratio  $X^{15}\text{N}/X^{14}\text{N}$  measured in any N-bearing species X is noted  $\mathcal{R}_X$ .

In our Solar System, any object (with the exception of Jupiter) is actually enriched in  $^{15}\text{N}$  compared to the PSN (see Fig. 1). Large excesses in  $^{15}\text{N}$  have been found in organic material of chondrites and Interplanetary Dust Particles (IDPs). Enrichments in  $^{15}\text{N}$  are measured at different scales of the material (bulk vs hotspots) and can be as high as  $\mathcal{R} = 50$  (Messenger, 2000; Bonal et al., 2010). Molecules in cometary coma also appear to be  $^{15}\text{N}$ -enriched, with  $\mathcal{R}$  ratios varying between 139 and 205 in HCN and CN (see the review by Jehin et al. (2009)).

The variation of the nitrogen isotopic composition in Solar System objects is most likely caused by a variety of effects. These

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**Fig. 1.** Nitrogen isotopic composition of Solar System objects as compared to the composition of simple molecules in interstellar clouds. The isotopic composition is expressed in term of  $^{14}\text{N}/^{15}\text{N}$  ratios (left scale) and in  $\delta^{15}\text{N}$  notation (right scale,  $\delta^{15}\text{N} = [R_{\oplus}/R - 1] \times 1000$ , where  $R_{\oplus} = 272$  is the nitrogen isotopic composition of the terrestrial atmosphere, see also Table 3). Square and circle symbols are for measurements made on molecules with amine and nitrile functional groups, respectively. IOM stands for insoluble organic matter, SOM for soluble organic matter, and CAI for calcium-, aluminum-rich inclusions. The range of values reported towards L183 and L1544 reflect the spatial variations across the sources.

include: (i) nucleosynthetic origin (Audouze, 1985; Adande and Ziurys, 2012, and references therein); (ii) photochemical self-shielding in the solar nebula (Clayton, 2002; Lyons et al., 2009); (iii) spallation reactions caused by the irradiation of the young Sun (Kung and Clayton, 1978; Chaussidon and Gounelle, 2006); and (iv) low temperature isotope exchanges (Terzieva and Herbst, 2000, hereafter TH00). The absence of large  $^{15}\text{N}$ -enrichments across the Galaxy (Adande and Ziurys, 2012, and references therein) and the small fractionation effects predicted by standard gas-phase chemical models (TH00) have weakened so far the hypothesis of a preserved (low temperature) interstellar chemistry to explain the  $^{15}\text{N}$ -enrichments observed in primitive solar cosmo-materials. However, the gas-grain chemical model of Charnley and Rodgers (2002) is able to reach a significant  $^{15}\text{N}$  enrichment of ammonia which is eventually locked into ices. The absence of a direct correlation between D and  $^{15}\text{N}$ -enrichments in organics from primitive cosmo-materials has also been interpreted as the lack of remnant interstellar chemistry for N isotopologues (Briani et al., 2009; Marty et al., 2010; Aléon, 2010).

In the present work, we analyze the line intensities of the fundamental rotational transitions of  $\text{H}^{13}\text{C}^{14}\text{N}$  and  $\text{H}^{12}\text{C}^{15}\text{N}$  ( $\text{H}^{13}\text{CN}$  and  $\text{HC}^{15}\text{N}$  in the following) towards two starless dense cores, L1544 and L183 (Hily-Blant et al., 2010). The main novelty in our analysis stems from the recent availability of accurate collisional hyperfine selective rate coefficients for HCN with  $\text{H}_2$  (Ben Abdallah et al., 2012). The present work (i) brings new observational constraints on nitrogen isotopic fractionation in gas phase and (ii) puts a new perspective on the actively debated and long questioning issue of the origin of the  $^{15}\text{N}$ -enrichments observed in primitive cosmo-materials as compared to the protosolar nebula.

## 2. Material and methods

### 2.1. Observations

Observations of the pure rotational  $J = 1-0$  lines of  $\text{H}^{13}\text{CN}$  and  $\text{HC}^{15}\text{N}$  were carried out with the IRAM-30m telescope by Hily-Blant et al. (2010). Spectra along perpendicular directions towards the L183 and L1544 starless cores were obtained, with extremely

high spectral resolution ( $\nu_0/\delta\nu \approx 4 \times 10^6$ ), such that the hyperfine structure of the  $\text{H}^{13}\text{CN}$  (1-0) is resolved. The details of the observational setup and hardware performances are available in Hily-Blant et al. (2010). The  $\text{H}^{13}\text{CN}$  and  $\text{HC}^{15}\text{N}$  (1-0) spectra towards L183 and L1544 are shown in Fig. A.4. The data are analyzed following a more robust method than the one previously adopted, where column densities were derived under the Local Thermal Equilibrium (LTE) assumption at a temperature of 8 K. In the present analysis, we make use of the hyperfine structure of the  $\text{H}^{13}\text{CN}$  (1-0) line and of new collisional coefficients for  $\text{HCN}-\text{H}_2$  (Ben Abdallah et al., 2012) which were also adopted for  $\text{H}^{13}\text{CN}$  and  $\text{HC}^{15}\text{N}$ .

### 2.2. Data analysis

The analysis of the data makes use of the hyperfine structure of  $\text{H}^{13}\text{CN}$ . The total opacity and excitation temperature of the  $\text{H}^{13}\text{CN}$  (1-0) transition are derived, assuming equal excitation temperature within the hyperfine multiplet. This assumption is justified as long as the opacity remains of the order of unity, which as will be seen later, holds for the lines towards L1544 and L183. The opacity and excitation temperature may then be used to derive the column densities under the LTE assumption (see details in the Appendix A). Alternatively, the opacity and line intensity may serve to compute the column density,  $\text{H}_2$  number density, and kinetic temperature, from non-LTE calculations, under the so-called large velocity gradient framework. In such case, we have used the RADEX public code (van der Tak et al., 2007). In these calculations, the  $\text{H}^{13}\text{CN}$  column density is searched for by varying the  $\text{H}_2$  density and the kinetic temperature in the range  $10^{11}-10^{14} \text{ cm}^{-2}$ ,  $10^3-10^7 \text{ cm}^{-3}$ , and 5–15 K, respectively.

In the case of L183, three methods have been compared. (1) The HFS method from the CLASS software was applied (see Appendix A) with the opacity and the excitation temperature as outputs, which in turn serve to compute a LTE column density. (2) Another fitting method was based on three independent Gaussians, yet constrained to have the same linewidth, whose peak intensities were used to derive the opacity and the excitation temperature. These two outputs give another LTE estimate of the total column density. (3) The opacity and line intensity of a given hyperfine component (e.g. the one with  $\text{RI} = 0.5556$ ) from the latter fitting method were

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