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Adsorption experiments of ammonia and clay minerals to understand nitrogen isotopic fractionation in molecular clouds

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Nitrogen is the fifth abundant element in the universe and also essential component of organic molecules. Various nitrogen-containing organic compounds have been found by laboratory analysis of extraterrestrial materials. The stable isotopic composition of nitrogen ($^{15}N/^{14}N$ ratio) will give information about evolutionary history of the organic molecules. Primitive solar system materials such as chondrites, comets, and interplanetary dust particles (IDPs) show various degrees of ^{15}N-enrichment compared to the solar system value of -400 ‰ [1]. They display up to +1500 ‰ in the bulk $\delta^{15}N$ value (‰, normalized as vs. AIR) [2, 3]. Furthermore, anomalously high ^{15}N-enrichments, as called hot spots, have been frequently found within a single material with the highest $\delta^{15}N$ values reaching as high as +5000 ‰ [4]. These ^{15}N-enrichments are considered to be originated in cold interstellar environments. However, the mechanisms of isotopic fractionation of nitrogen in the interstellar medium are not well understood and only a few models have been proposed [e.g., 5].

In this study, we focused on adsorption process of ammonia on grain surface of interstellar dusts as a potential mechanism for the extreme ^{15}N-enrichment and its high-heterogeneity found in extraterrestrial materials. Ammonia is a primitive nitrogen-containing compound and also one of major molecules in molecular clouds. Since ammonia is a highly reactive chemical, it is a precursor for nitrogen-involving organic molecules. The adsorption of ammonia on grain surface would be the first step for the formation of more complicated organic molecules. In order to examine the isotopic fractionation of nitrogen through adsorption of ammonia on grain surface, we performed experiments using ammonia gas and several adsorbents. For the experiments, six clay minerals (montmorillonite, saponite, dickite, kaolinite, pyrophyllite, and halloysite) were selected as the adsorbents. They were kept at 110 ℃ prior to the experiments to minimize adsorbed water. The each clay mineral was enclosed into a vacuumed glass vial and then ammonia gas (27 ‰, SI science) was introduced. A few days later, the glass vial was opened and the nitrogen isotopic composition of the adsorbed ammonia was determined by nanoEA/IRMS [6]. The results showed a relationship between $\delta^{15}N$ values and the adsorbed ratio, which is explained by Rayleigh fractionation model. The adsorbents with low adsorption ratio have higher $\delta^{15}N$ values compared to initial ammonia gas. The difference in the degree of ^{15}N-enrichment and adsorption property among clay minerals was also observed. These results imply that the adsorption of ammonia on grain surface should be considered as one of potential scenarios for ^{15}N-enrichment.

Reference: [1] Marty B. et al. (2011) Science 332, 1533. [2] Bonal L. et al. (2010) GCA 74, 6590. [3] Manfroid J. et al. (2009) A&A 503, 613. [4] Briani G. et al. (2009) PNAS 106, 105222. [5] Rodgers S.D. & Charnley S.B. (2008) Mon.Not.R.Astron.Soc.385, L48. [6] Ogawa et al. (2010) in Earth, Life, and Isotopes. pp.339.

Keywords: nitrogen isotopic fractionation, adsorption, ammonia, molecular clouds