Mass Dependency Of Isotope Fractionation Of Gases Under Thermal Gradient

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Thermal diffusion (TD) unmixes elements/isotopes of gas molecules under temperature gradient. Classical kinetic theory treats gas molecules as spherical and collisions being elastic and isotope separation is not pressure and temperature dependent, but composition dependent. In this sense, molecular mass is the essential factor for TD separation of isotopologues. Anomalous isotope fractionations, however, were observed half century ago, for N_2, CO, CO_2 and O_2 which violate the mass dependency of TD, e.g. the sign of isotope enrichment along the thermal gradient is opposite to the theoretical prediction. Recent reports by us also showed that the isotopologues of O_2 and SF_6 gases did not fractionate mass dependently and the deviation was both pressure and temperature dependent. Also, the SF_6 results showed no considerable S-36 anomaly, which suggests nuclear spin effect could be a responsible factor

We performed a series of experiments with thermal structures of diffusive and convective settings, on gases of Neon, O_2, CO_2 and CO. We used gases with natural-abundance isotopes in contrast with those with highly-enriched minor isotopes. Our results showed 1) three isotopologues for monatomic Neon are consistent with theoretical predicted mass relationship, suggesting nuclear spin is not a sufficient condition for the mass anomalies. 2) The anomalies persist in strong convections, suggesting TD could result in considerable isotope effect in natural conditions. 3) Anomalies are observed for all the polyatomic molecules, suggesting molecular structure and internal degree of freedom may play roles during TD in addition to the molecular masses.

Our observation is important for investigating intermolecular forces due to the seemingly strong dependence and sensitivity of multiple isotope signatures to the moleular properties and collision conditions; and for isotope partitioning associated with TD in the planetary upper atmosphere and stellar interiors as TD was suggested to be significant physical processes occurring in those environments.