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Mass Independent Isotopes In Nature

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With the 1983 discovery of a chemical process that alters stable isotopes in a mass independent manner, a new branch of isotope geochemistry was opened. In the ensuing years, there have been numerous efforts to provide and experimental and theoretical details of the mechanism responsible for the unique fractionation process. Though there remain areas of needed theory, a great deal has been resolved and the quantum mechanical basis is now understood in significantly greater detail. The measurements of these anomalous isotopic compositional patterns have in fact provided a new way to understand gas phase chemical reactions.

One of the most striking features of the first published work was that the oxygen isotopic fractionation pattern directly mimics that observed in the calcium aluminum inclusions of the Allende meteorite. Those specific isotopic compositions had previously been thought to arise from an astrophysical process, such as supernovae. The nuclear models have been abandoned and it is now generally accepted that chemical processes are responsible for production of the anomalies. There are several potential models in play at present, including chemical reactions and photochemical self shielding, both of which will be discussed in this talk and session.

At present, all of the oxygen bearing species in the atmosphere possess mass independent isotopic compositions. The specifics of these patterns provide information related to the oxidation/chemical transformation pathway as well as transport. Aerosol sulfate and nitrate measurements have been a powerful new means by which their biogeochemical cycles may be elucidated. Numerous environments, ranging from the South Pole, Indian Ocean, Chilean and Namibian deserts, trans- Pacific transport, and coastal California have been studied. This includes air, water, and soils.

Mass independent sulfur isotopic compositions in the Earth's earliest atmosphere have provided a new means by which the origin and evolution of oxygen and ozone may be studied, as well as to constrain their partial pressures. In addition, mass independent sulfur and oxygen isotopic measurements of Martian meteorites have provided a mechanism to understand the coupling of the Martian atmosphere to the regolith, and the formation of associated secondary minerals.