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Artifact-Free Measurements of Isotopic Composition for Atmospheric and Planetary Gas Analysis

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The isotopic composition of gaseous species can provide critical information regarding the age and chemical or physical origin of sample material. However, the challenge of maintaining isotope abundance scales – generated by comparing sample measurements to those of a reference material having finite quantity and stability – may limit inter-laboratory agreement and consequently the uncertainty evaluation of new measurement methods. Here we will present progress towards realization of artifact-free isotope scales and rapid measurements of isotopic composition enabled by absolute SI-traceable measurement schemes.

We will discuss how cavity ring-down spectroscopy techniques, capable of highly precise and accurate measurements of transition-resolved peak areas, can be leveraged in combination with quantum chemical calculations of transition moments to enable measurement of molecular isotopologue ratios [1]. We will also introduce direct frequency comb spectroscopy methods for rapid and precise measurement of isotopic abundance [2]. This discussion will include demonstrations in the near- and mid- infrared spectral regions employing cross-dispersed spectrometers. Implications for carbon, nitrogen, and oxygen isotopic analysis will be presented.

Applications of these SI-traceable measurement approaches include accurate source apportionment and greenhouse gas inventories, radiocarbon dating, isotope forensics, with the potential for high-impact contributions to emerging advances in exoplanetary studies and astrophysics.

[1] A. J. Fleisher, H. Yi, A. Srivastava, et al., *Nat. Phys.* **2021**, 17, 889-893

[2] D. M. Bailey, G. Zhao, and A. J. Fleisher, *Anal. Chem.* **2020**, 92 (20), 13759–13766