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New high-resolution isotope mass spectrometry applications for atmospheric sciences

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Stable isotope measurements have become an important scientific tool to constrain the budgets of atmospheric compounds. Because of analytical limitations, isotope investigations have generally targeted single substituted isotopocules for most species. Recently developed high-resolution, high-sensitivity isotope ratio mass spectrometers enable the measurement of multiply substituted isotopocules, and expand the possibilities to determine isotope ratios on fragment ions that are formed in the ion source of mass spectrometers. I will present four applications that were recently realized with the MAT 253 instrument at Utrecht University: Measurements of the clumped isotopes ($^{17}O^{18}O$ and $^{18}O^{18}O$) in atmospheric O_2, measurements of DD in atmospheric H_2, measurements of the clumped isotopic composition of CH_4 and measurements of the ^{17}O anomaly of CO_2 on O atom fragments. These signatures can generally be determined with a precision at or close to the counting statistics limit with the MAT 253. This means that the final measurement precision is determined by the signal intensity and observation time. Therefore, measurement time, sample size and instrument stability are the most important practical limitations for high-precision measurements. As higher mass resolution comes at the cost of lower signal intensity, the resolution is chosen for each gas to just allow separation of the most critical interferences. The presentation will mostly focus on the technical implementation of the clumped isotope techniques and their calibration.