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Chlorine isotope fractionation in stratospheric difluorodichloromethane (CFC-12)

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Previous isotope measurements in trace gases above the lowermost stratosphere have been restricted to a few substances only, mainly N_2O, CH_4, CO_2, O_3, H_2 and H_2O. This is because of the difficulty to obtain sufficient amounts of air for the analysis of less abundant gases. Conventional wisdom holds that the higher precision of dedicated multi-collector isotope ratio mass spectrometers is required to deduce meaningful information on budgets, chemistry and transport of trace gases from isotope measurements.

We challenge this perception and report on the first measurements of the chlorine isotope composition of stratospheric difluorodichloromethane (CFC-12) using quadrupole and single-collector mass spectrometers. Whole air samples from tropical balloon campaigns were analysed using standard GC-MS techniques. CF_2Cl_2 mixing ratios were between 50 and 500 pmol mol^{-1}. Similar to stratospheric N_2O and CH_4, we find an increase in the relative isotopic enrichment, (^{37}Cl), with altitude and a tight correlation between ln[1+ (^{37}Cl)] and ln(mixing ratio). The relative isotope ratio difference between the most enriched stratospheric sample and tropospheric air exceeds the entire range for (^{37}Cl) in terrestrial materials reported before this study.

Our results provide us with top-down constraints on the global CF_2Cl_2 budget. In combination with isotopic analysis of industrial CF_2Cl_2 sources, we will be able to obtain estimates of stratosphere-troposphere exchange.