## Triple oxygen isotope composition of CO<sub>2</sub> in the upper troposphere and stratosphere

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High precision measurements of  $\Delta'^{17}$ O can be used to constrain biosphere-atmosphere exchange of CO<sub>2</sub>, the residence time of tropospheric CO<sub>2</sub> and stratosphere-troposphere exchange. In this study, we report measurements of the  $\Delta'^{17}$ O(CO<sub>2</sub>) from air samples collected during two aircraft based programs, CARIBIC and StratoClim. CARIBIC (Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container) provided air samples from numerous transcontinental flights in the upper troposphere/lower stratosphere region. StratoClim (Stratospheric and upper tropospheric processes for better climate predictions) conducted intensive campaigns with the highaltitude aircraft M55 Geophysica during the Asian Summer Monsoon Anticyclone (ASMA), providing air samples from altitudes up to 21 km.

Using high precision  $\Delta'^{17}O$  measurements of the CARIBIC samples, we show that the N<sub>2</sub>O- $\Delta'^{17}O$  correlation, previously observed in the stratosphere, extends to the upper troposphere. Moreover, we found no significant spatial or hemispheric differences in  $\Delta'^{17}O(CO_2)$  for the upper tropospheric samples collected during the CARIBIC program. However, in many of the StratoClim samples, with significant stratospheric contributions, we observed a much shallower N<sub>2</sub>O- $\Delta'^{17}O$  slope compared to CARIBIC samples and previous publications. This deviation is attributed to active mixing between tropospheric and stratospheric air above the tropopause within the ASMA, confirming previously published model calculations. these samples provide the first experimental evidence that differences in vertical mixing led to significantly different N<sub>2</sub>O- $\Delta'^{17}O$  slopes, as suggested in previous model calculations. High precision  $\Delta'^{17}O$  measurements can identify ejections of tropospheric air into the stratosphere based on the slope of the N<sub>2</sub>O- $\Delta'^{17}O$  correlation, as both tracers have chemical lifetimes longer than their transport times.

Furthermore, we use the  $\Delta'^{17}O$  measurements from the lower stratosphere and the upper troposphere to estimate global stratospheric production and surface removal of the isotope tracer  $\Delta'^{17}O$ . The removal estimate is then used to derive an independent estimate of global vegetation exchange of CO<sub>2</sub>, confirming earlier estimates based on surface level  $\Delta'^{17}O$  measurements.