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# Long-term observation of mass-independent oxygen

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Abstract. Stratospheric and upper tropospheric air samples were collected during 1994–2004 over Sanriku, Japan and in 1997 over Kiruna, Sweden. Using these archived air samples, we determined the triple oxygen-isotope composition of stratospheric CO<sub>2</sub> and the N<sub>2</sub>O mixing ratio. The maximum  $\Delta^{17}O_{CO_{2}}$  value of +12.2‰, resembling that observed previously in the mesosphere at 60 km height, was found in the middle stratosphere over Kiruna at 25.6 km height, suggesting that upper stratospheric and mesospheric air descended to the middle stratosphere through strong downward advection. A least-squares regression analysis of our observations on a  $\delta^{18}O_{CO_2} - \delta^{17}O_{CO_2}$  plot (r<sup>2</sup>>0.95) shows a slope of 1.63±pm0.10, which is similar to the reported value of 1.71±0.06, thereby confirming the linearity of three isotope correlation with the slope of 1.6-1.7 in the mid-latitude lower and middle stratosphere. The slope decrease with increasing altitude and a curvy trend in three-isotope correlation reported from previous studies were not statistically significant. Using negative linear correlations of  $\Delta^{17}\text{O}_{\text{CO}_2}$  and  $\delta^{18}\text{O}_{\text{CO}_2}$  with the N<sub>2</sub>O mixing ratio, we quantified triple oxygen-isotope fluxes of CO<sub>2</sub> to the troposphere as +48‰ GtC/yr ( $\Delta^{17}\text{O}_{\text{CO}_2})$  and +38‰ GtC/yr ( $\delta^{18}\text{O}_{\text{CO}_2})$  with ~30% uncertainty. Comparing recent model results and observations, underestimation of the three isotope slope and the maximum  $\Delta^{17}\text{O}_{\text{CO}_2}$ value in the model were clarified, suggesting a smaller O2 photolysis

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