Triple oxygen composition of carbon dioxide from fossil fuel combustion

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The oxygen triple isotope composition of CO$_2$ from different natural and anthropogenic sources is gaining in importance as possible tracer of gross carbon exchanges between major reservoirs [1]. Determination of carbon dioxide isotope composition (given as isotope anomaly relative to the rocks and minerals defined terrestrial fractionation line: $\Delta^{17}$O$_{TFL}$) of different provenance is crucial for enhance atmospheric modelling. The isotope anomaly of CO$_2$ from fossil fuel combustion is especially interesting as it is the main form of anthropogenic carbon release. Here, we report the first data on the $\Delta^{17}$O of CO$_2$ from fossil fuel combustion.

CO$_2$ was collected above the flame of a propane-butane stove. The collected gas mixture was dried in a P$_2$O$_5$ water trap and the CO$_2$ was isolated from non-condensable gases with a Russian doll type cryogenic trap at -196 °C [2]. The amount of CO$_2$ was determined in a calibrated volume. The $\Delta^{17}$O$_{TFL}$ value of CO$_2$ inferred from oxygen isotope equilibration with CeO$_2$ at 685°C, and subsequent CeO$_2$ analysis by means of IR laser fluorination GC-CF-irmMS [3,4]. The amount of carbon dioxide was approx. 2% in the exhausted gas.

We determined a $\Delta^{17}$O$_{TFL}$ value of the CO$_2$ of $-0.52 \pm 0.02$‰ (relative to a TFL with $\beta_{TFL} = 0.525$). The $\delta^{18}$O$_{SMOW}$ of the CO$_2$ was $+22$‰. The CO$_2$ is carrier of a considerable negative isotope anomaly. The anomaly is larger in magnitude than the anomaly of tropospheric air O$_2$ $-0.388 \pm 0.032$‰ [5].

Either formation of CO$_2$ during combustion of propane in air is accompanied by a mass-independent isotope effect or other processes affected the resultant $\Delta^{17}$O of CO$_2$. Such effects could include (partial) equilibration with H$_2$O and/or kinetic fractionation during combustion with air O$_2$.

In ongoing work, we will investigate CO$_2$ from different combustion processes, like in diesel and petrol engines, natural gas heating systems and burning of firewood. Along with CO$_2$, we will also analyse the triple oxygen isotope composition of H$_2$O produced during combustion.