Variations of atmospheric CO2 and its isotopes in the upper troposphere/lower stratosphere over Siberia.

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Abstract:
Carbon and oxygen isotope ratios ($\delta^{13}C$ and $\delta^{18}O$) of atmospheric CO2 provide us useful information on an understanding of the global carbon cycle owing to their dependencies on sources or sinks. To reveal temporal and spatial variations of $\delta^{13}C$ and $\delta^{18}O$ of CO2 in the upper troposphere/lower stratosphere (UT/LS) over Siberia, we analyzed air samples collected on board commercial airliners between France/Russia and Japan by the Comprehensive Observation Network for TRace gases by AIrLiner (CONTRAIL) project. The CO2 mixing ratio and $\delta^{18}O$ show a secular increase during the observation period from April 2012 to May 2017. In contrast, $\delta^{13}C$ shows a secular decrease mainly due to isotopically lighter CO2 emissions by fossil fuel combustion. The observed change rate of $\delta^{13}C$ with respect to CO2 for the long-term trend (-0.02 ‰ ppm$^{-1}$) is, however, significantly smaller than that expected from fossil fuel combustion (-0.05 ‰ ppm$^{-1}$). This is ascribed to an isotopic disequilibrium of CO2 between the atmosphere and the oceans or the terrestrial biosphere. The negative correlation between the CO2 mixing ratio and $\delta^{13}C$ is also observed in the seasonal timescale variations in the UT. The change rate of -0.043 ‰ ppm$^{-1}$ suggests that they are seasonally driven by the carbon exchange between the terrestrial biosphere and the atmosphere. On the other hand, those in the LS are primarily governed by seasonal cycle in stratosphere-troposphere exchange (STE) processes including the Brewer-Dobson circulation (BDC) in the stratosphere. A similar involvement with the STE process is also found in the seasonal relationship between CO2 and $\delta^{18}O$; in particular, it is crucial that deeper stratospheric air masses with low CO2 and high $\delta^{18}O$ influenced by stratospheric ozone are transported down to the UT/LS region in winter/spring in association with the BDC.