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On the use of N_2O as a tracer of stratosphere to troposphere flux of long-lived trace gases based on CARIBIC data.

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Stratosphere-troposphere exchange (STE) plays an important role in the distribution of several trace gases, with ozone and CO being commonly in use to study STE mixing structures and processes. Here we explore the use of N_2O as a tracer of STE mixing proportions and the STE flux for long-lived gases, in particular CO_2 and CO_2 isotopes. N_2O has a near uniform distribution in the troposphere, with little seasonal variation and the only sink in the stratosphere. Between 2005 and the present measurements of N_2O in flasks collected as part of the CARIBIC project have resulted in a large data set with excellent N_2O accuracy. N_2O data for the UTLMS at mid- and high-latitudes (40-77 deg N) are found to demonstrate highly systematic STE mixing trends and compact distributions of the tropospheric end-member.

First, upper-tropospheric and STE-affected air can easily be distinguished using an N_2O threshold based on the N_2O distributions at NOAA stations. The threshold value is found to be the same for mid- and high- latitudes, with the data uncertainty being close to the seasonality. Second, as the UTLMS at mid-latitudes may contain both stratospheric air and a significant fraction of polar vortex air having experienced different degrees of chemical ozone losses, N_2O can be used as a linear /additive tracer for long-lived gases, in particular when data are to be compared over different seasons and/or latitudes. Third, CARIBIC CO_2 isotope data for the UTMLS are found to agree well with the d18O(CO_2)/N_2O slope estimated based on published stratospheric CO_2 isotope data, thus supporting our understanding of STE mixing and linear additive identity for N_2O. We conclude that CARIBIC data coupled with N2O and CO_2 isotopic data published for the stratosphere and stations can be used as the frame of reference for d18O(CO_2) and D17O(CO_2) STE fluxes. The latter is of particular interest as it is thought to constrain global gross CO_2 fluxes. Our data can also help trace STE flux by deep stratospheric intrusions e.g. at mountain stations, an example of which will also be presented at the meeting.