Geophysical Research Abstracts
Vol. 13, EGU2011-13243, 2011
EGU General Assembly 2011

Source emission signals in variabilities of nitrous oxide and methane in the upper troposphere over the western Pacific deduced from tagged tracer simulations

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Multiple greenhouse gases simulations with tagged-tracers are performed to mainly understand surface-source influences on latitudinal-temporal variabilities of nitrous oxide (N2O) and methane (CH4) in the upper troposphere (UT) over the western Pacific observed by the Automatic air Sampling Equipment (ASE) in the CONTRAIL project. We use greenhouse gases concentration data, which have been obtained almost fortnightly in the altitude range of 9-11 km between Sydney or Brisbane, Australia and Tokyo, Japan for the period Dec 2005 - Mar 2009. An atmospheric general circulation model-based chemistry transport model (ACTM), which is nudged toward the Japanese 25 year ReAnalysis data from the Japan Meteorological Agency (JMA) (JRA-25), is used in this study. Dynamical structure in the tropical UT region in ACTM is reasonably validated by the fact that ACTM simulation of mean latitudinal SF6 gradient for the observation period is almost perfectly consistent with the observation within 0.03ppt, which is small enough compared to the measurement precision. For tracing the origins of N2O and CH4 in model, the globe is separated into more than ten of emission regions so that each region's emission affects the corresponding tagged-tracer's concentration variation on the globe through the atmospheric transport. The temporal variations of tracers simulation results and observation results are detrended, and their seasonal or shorter eventual concentration variabilities are compared. In case of N2O, concentration values around 30N and 30S are largely fluctuated by stratospheric intrusions, which lower N2O concentration in the UT, but some surface source signals can be still detectable especially around 30N by removing data highly affected by the stratosphere. That indicates that N2O around 20-30N seems to be most affected by Middle East and South Asia emission region, and secondly by East and Southeast Asia emission region mainly in boreal summer. It is possible that monsoon and Tibetan Plateau work to transport N2O emitted from South Asia to this region in the UT over the western Pacific. Around 10S, N2O seems to be relatively dominated by emissions from Australia, but the degree is not so prominent compared to above two Asian regions for 20-30N. CH4 simulations show slightly different features of affecting source regions from those of N2O. It is indicated that China and India affect 26-30N region in the UT over the western Pacific in boreal summer and fall, respectively. Meanwhile, CH4 variations around 10S are dominantly affected by Malaysia-Indonesia-Papua New Guinea emission region. Difference of source regions affecting this region (around 10S) between N2O and CH4 might be related to difference of atmospheric lifetime in this region, because CH4 is actively broken by reaction with OH, which is relatively enhanced in the tropics, while N2O has no chemical loss in the troposphere.