

Nitrogen oxides in the global upper troposphere interpreted with cloud-sliced NO₂ from the Ozone Monitoring Instrument

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Nitrogen oxides ($\text{NO}_x \equiv \text{NO} + \text{NO}_2$) are long lived in the upper troposphere (UT), and so have a large impact on ozone formation where ozone is a powerful greenhouse gas. Measurements of UT NO_x are limited to summertime aircraft campaigns predominantly in North America. There are year-round NO_x measurements from instruments onboard commercial aircraft, but NO_2 measurements are susceptible to large interferences. Satellites provide global coverage, but traditional space-based NO_2 observations only provide one piece of vertical information in the troposphere. New cloud-sliced satellite NO_2 products offer additional vertical information by retrieving partial NO_2 columns above clouds and further exploit differences in cloud heights to calculate UT NO_2 mixing ratios. Two new cloud-sliced NO_2 products from the Ozone Monitoring Instrument (OMI; 2004 launch) provide seasonal UT NO_2 data centered at 350 hPa for 2005-2007 (NASA product) and 380 hPa for 2006 only (KNMI). Differences between the products include spectral fitting to obtain NO_2 along the viewing path (slant column), the air mass factor calculation to convert slant columns to true vertical columns, treatment of the stratospheric NO_2 component, and the choice of cloud products. The resultant NASA NO_2 mixing ratios are 30% higher than KNMI NO_2 and are consistent with summertime aircraft NO_2 observations over North America. Comparison between NASA NO_2 and the GEOS-Chem chemical transport model exposes glaring inadequacies in the model. In summer in the eastern US lightning NO_x emissions are overestimated by at least a factor of 2, corroborated by comparison of GEOS-Chem and MOZAIC aircraft observations of reactive nitrogen (NO_y). Too fast heterogeneous hydrolysis of dinitrogen pentoxide (N_2O_5) leads to an underestimate in UT NO_2 in winter across the northern hemisphere. Absence of interannual variability in lightning flashes in the lightning NO_x parameterization induces biases in UT NO_2 in the tropics due to anomalous lightning activity linked to the El Niño Southern Oscillation. Ongoing work is to use GEOS-Chem to investigate the implications of updated representation of UT NO_x on ozone.