Nitrogen oxides in the global upper troposphere interpreted with cloud-sliced NO$_2$ from the Ozone Monitoring Instrument

Eloise A Marais (1), Daniel J Jacob (2,3), Sungyeon Choi (4), Joanna Joiner (4,5), Maria Belmonte-Rivas (6), Ronald C Cohen (7,8), Thomas B Ryerson (9), Andrew J Weinheimer (10), and Andreas Volz-Thomas (11)


Nitrogen oxides (NO$_x$ ≡ NO + 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$_2$O$_5$) 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.