



Nitrogen oxides in the global upper troposphere interpreted with cloud-sliced NO₂ 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)

(1) University of Birmingham, School of Geography, Earth and Environmental Sciences, Birmingham, UK, (2) John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge, MA, (3) Earth and Planetary Sciences, Harvard University, Cambridge, MA, (4) Science Systems and Applications Inc., Lanham, MD, (5) NASA Goddard Space Flight Center, Greenbelt, MD, (6) Royal Netherlands Meteorology Institute, De Bilt, the Netherlands, (7) Department of Chemistry, University of California at Berkeley, Berkeley, CA, (8) Department of Earth and Planetary Science, University of California at Berkeley, Berkeley, CA, (9) Chemical Sciences Division, Earth System Research Lab, National Oceanic and Atmospheric Administration, Boulder, CO, (10) National Center for Atmospheric Research, Boulder, CO, (11) IAGOS-AISBL, Brussels, Belgium

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.