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Rapid increases in tropospheric ozone over China observed by TES and attributed with OMI and TM5

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Tropospheric ozone is an important global air pollutant originating from photo-chemical oxidation of precursors such as volatile organic compounds (VOCs) and CO in the presence of NO_x, and from stratosphere-troposphere ozone exchange and horizontal transport. Tropospheric ozone has likely been increasing over the last 30 years but trends have flattened over the last decade. There are some indications that eastern Asia may be the exception to this rule, as this region is witnessing fast-growing ozone precursor emissions. Assessing ozone trends in the troposphere is difficult due to scarcity of long-term measurement sites and because of the relatively short lifetime of ozone and the influence of meteorology. Spaceborne sensors are characterized by their extensive spatial coverage and robust retrievals and are thus excellent tools to map spatio-temporal patterns in tropospheric ozone. In this study we evaluate time series of tropospheric ozone observed from space by TES (Tropospheric Emission Spectrometer onboard NASA's EOS-Aura satellite) with the TM5 chemical transport model (CTM) using seven years (2005-2011) of observations and model simulations for eastern Asia.

We show a clear increase of ozone mixing ratios in the free troposphere (FT) of about 9 ppbv at 464 hPa over eastern China between 2005 and 2011 using TES observations. This is a significant (p-value < 0.05) yearly increase of +1.33 ppbv based on weighted linear regression. To better understand what drives the tropospheric ozone increases, a TM5 reference run - using ECMWF meteorological fields and fixed RETRO & REAS anthropogenic emission inventories for the year 2006 - was conducted for the period 2005-2011 and compared with the TES ozone measurements. TM5 data were sampled at TES overpass times and the TES averaging kernel was applied on TM5 in order to conduct a proper evaluation. The TM5 simulations with constant 2006 NO_x emissions reproduce part of the observed increase in FT ozone (+0.78 ppbv/yr). We use OMI NO_2 observations to infer a more realistic magnitude and temporal pattern in the NO_x emissions over China. By implementing these improved NO_x emission estimates in the TM5 model, we obtain much better agreement for the simulated NO_2 columns as well as the ozone concentrations with EOS-Aura observations. The TM5 simulations now indicate a trend in FT ozone of +1.09 ppbv/yr for 2005-2011 over China, which is more consistent with the trend in the TES data. We will discuss the effect of stratospheric inflow and horizontal transport on the ozone trends observed in China.