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Remote sensed and in situ constraints on processes affecting tropical tropospheric ozone

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Abstract. We use a global chemical transport model (GEOS-Chem) to evaluate the consistency of satellite measurements of lightning flashes and ozone precursors with in situ measurements of tropical tropospheric ozone. The measurements are tropospheric O_3, NO_2, and HCHO columns from the GOME satellite instrument, lightning flashes from the OTD and LIS satellite instruments, profiles of O_3, CO, and relative humidity from the MOZAIC aircraft program, and profiles of O_3 from the SHADOZ ozonesonde network. We interpret these multiple data sources with our model to better understand what controls tropical tropospheric ozone. Tropical tropospheric ozone is mainly affected by lightning NO_x and convection in the upper troposphere and by surface emissions in the lower troposphere. Scaling the spatial distribution of lightning in the model to the observed flashes improves the simulation of O_3 in the upper troposphere by 5–20 ppbv versus in situ observations and by 1–4 Dobson Units versus GOME retrievals of tropospheric O_3 columns. A lightning source strength of 6±2 Tg N/yr best represents in situ observations from aircraft and ozonesonde. Tropospheric NO_2 and HCHO columns from GOME are applied to provide top-down constraints on emission inventories of NO_x (biomass burning and soils) and VOCs (biomass burning). The top-down biomass burning inventory is larger than the bottom-up inventory by a factor of 2 for HCHO and alkenes, and by a factor of 2.6 for NO_x over northern equatorial Africa. These emissions increase lower tropospheric O_3 by 5–20 ppbv, improving the simulation versus aircraft observations, and by 4 Dobson Units versus GOME observations of tropospheric O_3 columns. Emission factors in the a posteriori inventory are more consistent with a recent compilation from in situ measurements. The ozone simulation using two different dynamical schemes (GEOS-3 and GEOS-4) is evaluated versus observations; GEOS-4 better represents O_3 observations by 5–15 ppbv, reflecting enhanced convective detrainment in the upper troposphere. Heterogeneous uptake of HNO_3 on aerosols reduces simulated O_3 by 5–7 ppbv, reducing a model bias versus in situ observations over and downwind of deserts. Exclusion of HO_2 uptake on aerosols increases O_3 by 5 ppbv in biomass burning

regions, reducing a model bias versus MOZAIC aircraft measurements.

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